Naturally Occurring Radioactive Material (NORM VI)

Proceedings
of an International Symposium
Marrakesh, Morocco, 22–26 March 2010

IAEA
International Atomic Energy Agency

NATURALLY OCCURRING RADIOACTIVE MATERIAL
(NORM VI)

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PROCEEDINGS SERIES

NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM VI)

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NATURALLY OCCURRING RADIOACTIVE MATERIAL
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THE CADI AYYAD UNIVERSITY OF MARRAKESH
IN COOPERATION WITH
THE INTERNATIONAL ATOMIC ENERGY AGENCY
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FOREWORD

All minerals and raw materials contain radionuclides of natural origin, of which the most important for the purposes of radiation protection are the radionuclides in the ^{238}U and ^{232}Th decay series and ^{40}K. For most human activities involving minerals and raw materials, the levels of exposure to these radionuclides are not significantly greater than normal background levels. Such exposures, while having been the subject of much research, are not of concern for radiation protection. However, certain work activities can give rise to significantly enhanced exposures that may need to be controlled by regulation. Material giving rise to these enhanced exposures has become known as naturally occurring radioactive material (NORM).

Historically, most regulatory attention has been focused on the mining and processing of uranium ore, because such activities are a direct consequence of the radioactivity in the ore and form part of the nuclear fuel cycle. Over the past decade or two, however, more and more countries have introduced measures to regulate exposures arising from a wider range of natural sources, in particular minerals and raw materials other than those associated with the extraction of uranium. Two important developments in this regard were the establishment of the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (published in 1996 as IAEA Safety Series No. 115) and the European Council Directive 96/29/Euratom of 13 May 1996, both of which contained provisions for protective measures against significantly increased exposures of workers and members of the public to natural sources.

As a direct consequence of the European Council Directive and its possible implications for non-nuclear industries in Europe, a symposium on NORM, the first in the current series, was held in Amsterdam in 1997. The second in the series (NORM II) was held in 1998 in Krefeld, Germany, the third (NORM III) in Brussels in 2001, the fourth (NORM IV) in Szczyrk, Poland, in 2004 and the fifth (NORM V) in Seville, Spain, in 2007. In addition, a symposium on Technologically Enhanced Natural Radiation was held in Rio de Janeiro in 1999, reflecting the growing interest within regions beyond Europe in the management of exposure to NORM. The close involvement of the IAEA in most of these symposia is reflected in the fact that the proceedings of the Rio de Janeiro and Szczyrk symposia have been published as IAEA-TECDOC-1271 and IAEA-TECDOC-1472, respectively, while the Seville symposium was published in the IAEA Proceedings Series.

In the case of this NORM VI symposium, the IAEA entered into a formal cooperation arrangement with the organizing bodies, the Hassan II University of Mohammedia and the Cadi Ayyad University of Marrakesh, in terms of which the IAEA, in addition to publishing these Proceedings, served on the Steering

Committee and Scientific Committee of the symposium and provided financial support to several participants from Member States eligible to receive assistance under the IAEA technical cooperation programme. The Hassan II University of Mohammedia and the Cadi Ayyad University of Marrakesh were assisted in the organization of the symposium by two other Moroccan institutions, the Centre National de l’Energie, des Sciences et des Techniques Nucléaires (CNESTEN) and the Centre d’Etudes et de Recherches des Phosphates Minéraux (CERPHOS).

The NORM VI symposium was attended by 140 participants from 33 countries and provided an important opportunity to review the developments that had taken place during the three year period since the Seville symposium in 2007. This period, which began with the publication of new radiation protection recommendations by the International Commission on Radiological Protection, was characterized by ongoing activities to revise international standards on radiation protection and safety. These Proceedings contain all 38 papers accepted for oral presentation and four rapporteur reports, as well as a summary that concludes with the main findings of the symposium. Text versions of 43 poster presentations are provided on a CD-ROM which accompanies these Proceedings.

The IAEA, on behalf of the organizers, the Hassan II University of Mohammedia and the Cadi Ayyad University of Marrakesh, gratefully acknowledges the cooperation and support of all the organizations and individuals that have contributed to the success of this symposium. The IAEA officer responsible for this publication was D. Wymer of the Division of Radiation, Transport and Waste Safety.

EDITORIAL NOTE

The papers in these Proceedings (including the figures, tables and references) have undergone only the minimum copy editing considered necessary for the reader’s assistance. The views expressed remain, however, the responsibility of the named authors or participants. In addition, the views are not necessarily those of the governments of the nominating Member States or of the nominating organizations.

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SUMMARY

1. BACKGROUND TO THE SYMPOSIUM

1.1. Objectives

This symposium, the sixth in a series of symposia on naturally occurring radioactive material (NORM), once again provided an important opportunity to review recent technical and regulatory developments concerning exposure to NORM. The symposium brought together experts from a wide range of countries to report on and discuss the progress made in identifying, quantifying and managing the radiological risks associated with industrial processes involving NORM. The revision of international standards, which was presently in its final stages, provided an important backdrop to the presentations and discussion.

As with previous symposia in this series, the technical programme was well-subscribed, with 81 contributions being accepted for presentation. Nearly half of these were accepted for oral presentation and the balance in the form of posters. To help realize the objectives of the symposium, arrangements were made in the programme for each day’s presentations and discussions to be reviewed and summed up by a rapporteur. On the final day of the presentations, the rapporteur’s report included a review of the entire symposium and of the extent to which the objectives of the symposium were met.

1.2. International aspects

The first NORM symposium, held in Amsterdam, Netherlands in 1997, had been organized in response to concerns within the non-nuclear industry in the European Union (EU) that the implementation of a new European Council Directive (Directive 96/29/Euratom of 13 May 1996, laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation) would place unreasonable and unwarranted legal obligations on many industrial enterprises that handled and processed material containing low levels of radionuclides of natural origin. Subsequently, as new regulations for the control of exposure to NORM became established in EU Member States and as knowledge about levels of exposure improved, those concerns diminished to some extent, although the definition of the scope of regulation remained controversial. Furthermore, it became apparent that this was becoming more of a global issue because of the increasingly international profile of the mining and minerals processing industry, with large quantities of minerals being mined and beneficiated in countries remote from

Europe and shipped to other countries — often over vast distances — for further processing. In line with this trend, successive NORM symposia began to take on a more international flavour and the involvement of the IAEA became progressively greater. It is significant to note in this regard that NORM VI was the first in this series of symposia to be held outside Europe.

Given this background, it was decided that the planning of the NORM VI symposium should follow an approach similar to that adopted for NORM V in 2007, in that specific steps should be taken to encourage strong participation from countries outside the EU. The steering committee arranged for broad international representation on the scientific committee of the symposium and encouraged the members of that committee to actively promote participation in the symposium from within their own geographic regions. Furthermore, the IAEA provided financial support to nine participants from Member States eligible to receive assistance under the IAEA technical cooperation programme. Those efforts were evidently successful in that the symposium attracted 140 participants from 33 countries, including 18 countries outside the EU.

2. RADIATION PROTECTION STANDARDS AND REGULATORY APPROACHES

2.1. Revision of international standards

The keynote address in the opening session of the symposium described the progress made in revising the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS), which were published by the IAEA in 1996. Following a review in 2005 and 2006, a decision had been made to embark on a revision process starting in 2007 (the same year as NORM V, the previous symposium in this series) in collaboration with the cosponsoring organizations. That revision was now in its final stages. One of the most significant developments influencing the revision process was the publication, in 2007, of new radiation protection recommendations by the International Commission on Radiological Protection (ICRP). The drafting of the revised BSS has taken those new recommendations into account. In particular, the ICRP's categorization of exposure situations into three types — planned exposure situations, emergency exposure situations and existing exposure situations — is reflected in the draft document. Furthermore, the stringency of protection in existing exposure situations (referred to in the present BSS as chronic exposure situations) has been significantly increased by strengthening, and widening the scope of application of, the requirements for optimization of protection. In the case of radon, the stringency of control is further increased as a result of a recent statement by the ICRP in which it considers the health risk due to inhalation of radon to be significantly higher than previously assumed. Another important development in the revision process has been the greater use of quantitative criteria for deciding on the mechanism of control to be applied to exposures to natural sources and — in the case of exposures to be controlled as practices — for deciding on exemption and clearance.

2.2. Regulatory approach

It was clear from some of the presentations that governance structures at national and regional levels were important factors in implementing radiation protection standards and establishing regulatory systems. Countries such as Australia and the United States of America have federal systems of government involving relatively independent regulatory jurisdictions at different levels. The situation in the European Union was somewhat similar, in that individual Member States enacted their own legislation and regulations, but in overall compliance with legally binding Directives established at the European level.

A presentation on the regulatory approach to NORM in the USA highlighted some convoluted issues related to jurisdiction and terminology. The regulatory approach involves a mixture of federal and State regulation and was described as ‘uneven and fragmented’. The sytem of regulating radioactive materials is both qualitative and quantitative, being based on a radionuclide’s elemental identity and activity concentration, as well as its origin and processing history. The regulatory approach differs among the various jurisdictions and also differs from international standards. It was concluded that the prospect of a uniform and harmonized regulatory scheme is still only a hope for the future and that the current uncertainty with respect to NORM regulation in the USA will remain for the time being.

In Australia, the situation with regard to jurisdiction was clearer and did not appear to suffer the same problems as those in the USA. The agency responsible for radiation protection at the federal (Commonwealth) level only regulates Commonwealth entities and contractors and has no jurisdiction within individual States and Territories. In the past, however, the legislation and regulations in the various jurisdictions were not fully in place for all types of NORM activity and even now there are differences in legislation and regulations between the various jurisdictions. Since 2003, Australia has been involved in a process to provide NORM management guidance at the national level. After consultation with a wide variety of stakeholders, a Safety Guide has been developed that includes guidance on radiological issues in NORM management, operational issues, remediation of legacy sites and separate annexes on specific industries. The

Safety Guide is based on actual radiological data gathered for various NORM industries and follows very closely the most recent international standards published by the IAEA, including the adoption of a graded approach to the regulation of NORM based on the assessed level of risk to the environment, workers and members of the public and the concepts of exclusion, exemption and clearance.

The European Commission (EC) reported that it was actively participating in the IAEA's process for the revision of the BSS and, in parallel with that activity, was currently in the process of recasting five Euratom Council Directives (including Council Directive 96/29/Euratom referred to in Section 1.1) with the aim of merging them into a single Directive dealing with radiation safety. The aim was to achieve a high degree of harmonization between the revised BSS and the new European Council Directive. The draft European Council Directive contains new requirements for the control of exposure to natural sources, including specific requirements for building materials. Industrial activities involving NORM that are known to require regulatory consideration are listed. These activities are essentially the same as those identified in IAEA Safety Reports Series No. 49: Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials. A list of building materials of concern is also included. The draft Directive differs in some respects to the new BSS draft with regard to how the ICRP recommendations have been translated into regulatory requirements, but a substantial degree of overall consistency is evident.

2.3. Criteria for regulatory control of NORM

It was clear that, since the previous NORM symposium in 2007, many countries had made considerable progress in implementing radiation protection measures for the control of exposure to NORM. Increasing use was being made of international guidance published by the IAEA. In particular, the regulatory criterion of 1 Bq/g for the activity concentration of radionuclides in the uranium and thorium decay series was being accepted more widely in one form or another as a straightforward and pragmatic way forward. The principle behind this criterion was simply that the regulation of materials (other than possibly building materials) with activity concentrations within the normal range of environmental levels (0–1 Bq/g for uranium and thorium decay series radionuclides) was not a sensible use of regulatory resources.

There have been concerns that the adoption of the 1 Bq/g criterion could lead to unregulated situations in which members of the public could receive unacceptably high doses, particularly via contamination of groundwater used for drinking purposes, although such concerns have seemingly not been backed up by way of published literature. An investigation of this issue, conducted on behalf of the IAEA, had recently been completed and the results were presented at this symposium. The investigation focused on a member of the public living next to a 2 million t mineral residue deposit containing uranium and/or thorium decay series radionuclides each at an activity concentration of 1 Bq/g. The dose assessment was conducted using evidence based parameters published in the literature for a variety of real life situations involving large mineral residue deposits. The key conclusion of this investigation was that, for a conservative but nevertheless realistic exposure scenario, the maximum dose received by an individual, whether an adult or a young child, was unlikely to exceed about 0.2 mSv in a year. This finding is important because it underpins the decision to include the 1 Bq/g criterion in the current BSS draft — until now this criterion has appeared only as a guidance level in published IAEA standards.

For material (other than basic commodities such as food, water, fertilizer and building material) in which the activity concentration of any uranium or thorium decay series radionuclide exceeds 1 Bq/g, the current draft of the revised BSS requires that it be regulated as a practice and, accordingly, subject to the requirements for what are now referred to as planned exposure situations. In terms of the IAEA Safety Glossary, this material then falls within the definition of radioactive material and, more specifically, NORM. In applying the graded approach to regulation, the first step is to consider whether exemption is the optimum regulatory option. The current BSS draft specifies an annual dose of the order of 1 mSv as an appropriate criterion for exemption without further consideration, a value consistent with the natural background level and contrasting with the dose criterion of the order of 10 μSv for radionuclides of artificial origin (for which the natural background is essentially zero). Many symposium participants reported that this 1 mSv exemption level was being implemented already, at least for workers, and there was a clear impression gained that activities involving minerals and natural raw materials did not warrant the use of regulatory resources if they were unlikely to give rise to doses above this value. This same value of 1 mSv was also gaining general acceptance as a reference level for the use of commodities containing elevated levels of radionuclides of natural origin, particularly building materials.

It was clear from many of the presentations that a pragmatic, graded approach to the regulation of NORM was becoming increasingly accepted as a regulatory policy. For instance, there were reports from four countries (Sweden, Netherlands, Norway and Japan) that full regulation of NORM was applied only if the activity concentrations of uranium and thorium series radionuclides exceeded 10 Bq/g, with less rigorous forms of control being applied when the activity concentrations were between 1 and 10 Bq/g.

2.4. ICRP Task Group on NORM

At the conclusion of the symposium, a representative of the ICRP elaborated on the proposed activities of an ICRP Committee 4 Task Group on Application of the Commission’s Recommendations to NORM. The Task Group has been established to develop a decision aiding framework for the practical implementation of the Commission’s latest recommendations on radiation protection for NORM (as set out in ICRP Publication 103 in 2007). The framework would allow radiation protection principles to have practical and consistent inputs for the regulatory programmes implemented for protection of workers, the public and the environment. It was explained that the framework would cover the entire range of NORM activities, including shipment and waste management of bulk quantities, as well as the presence of NORM in consumer products, particularly construction materials. The Task Group would also seek to illustrate how the framework would be applied to certain activities that are currently of concern, such as oil and gas production, burning of coal and the production of rare earths and phosphate fertilizers. Recent publications and documents of other international organizations such as the IAEA and the EC would be taken into account.

The Task Group was seeking input on a variety of exposure situations, ranging from those that should be excluded from any control, to existing exposure situations where exposures were not significantly different from background exposures, up to planned exposure situations where occupational exposures may be of concern. It was also seeking input on questions such as: What are the important considerations in deciding whether or not to impose regulatory control? How can protection be optimized such that resources are not deflected away from general occupational health and safety or that an unwarranted burden is placed on existing industries? More specifically, the following topics were identified as needing consideration:

(a) The identification of problem areas where exposures to natural sources are present, particularly in industrial situations where radionuclide concentrations become enhanced in products or waste streams;
(b) The identification of any changes that might be needed in the ICRP system of protection relevant to NORM and how such changes would impact on NORM industries in terms of the management of exposures;
(c) The application of the concepts of existing and planned exposure situations, categories of exposed persons (public and occupational) and principles of exclusion and exemption;
(d) The identification of the ranges of activity concentrations and pathways that arise from NORM activities;

(e) A categorization of exposures into those that are within the normal range, those that occur at the higher end of the distribution and those in certain industries that could be regarded as unwarranted;
(f) An examination of the important pathways by which significant exposures can occur;
(g) The identification of those exposures that should be excluded or exempted from regulation;
(h) The identification of situations that could be classified as either existing exposure situations or planned exposure situations and recommendations on how such exposures should be optimized using, as appropriate, reference levels or dose constraints;
(i) Any overall conclusions or generalized recommendations.

2.5. General conclusions on safety standards and the regulation of NORM

Various important conclusions on standards and regulation emerged during the course of the symposium, including several related to the topics identified by the ICRP (see Section 2.4). One general conclusion was that some progress had been made in the three years since NORM V towards establishing a harmonized regulatory approach worldwide. However, there were still many examples of differences in standards and regulatory approaches between countries, and even within individual countries.

2.5.1. Determining the scope of regulation

Overall, there appeared to be a growing consensus on the identification of those particular industrial processes (and residues from those processes) that were most likely to require regulation as practices. However, there was still a tendency in some cases to adopt a very conservative and cautious approach, resulting in undue attention being given to industrial processes and residues for which there was no real evidence of the need for control. This could defeat the purpose of the exercise, which is to focus regulatory attention on those situations where it was most needed. In the same vein, several speakers referred to the need for an evidence based approach to the making of policy and regulatory decisions rather than depending on questionable risk assessments derived from conservative modelling and implausible exposure scenarios.

2.5.2. Interpretation of ICRP recommendations

Concerning the revision of international standards and the European Directive, it was apparent that both revision processes had had to contend with problems of interpretation of the 2007 recommendations of the ICRP, particularly concerning the distinction between existing exposure situations and planned exposure situations. These problems had given rise to many of the differences between the draft of the revised BSS and that of the proposed new European Council Directive. While it was felt that these differences would not necessarily lead to serious problems, there was concern that the ICRP had not fully thought through the implications of its 2007 recommendations for exposure to natural sources. For this reason, the establishment of the ICRP Task Group, while seen as being very much overdue, was nonetheless welcomed.

One general conclusion was that the distinction between existing exposure situations and planned exposure situations (referred to in current standards as intervention and practices, respectively) could not always be readily applied to exposure to natural sources and that attempts to do so had in some cases added to the confusion. In this regard it was noted that in Japan guidance developed recently for the control of exposure to NORM involves, for certain types of material, a combination of measures for both ‘practices’ and ‘intervention’. With the introduction of the new ‘exposure situation’ terminology in the 2007 ICRP recommendations, the problem appears to have been exacerbated because of the temptation to interpret the words ‘planned’ and ‘existing’ according to their literal English meanings, leading to further confusion rather than clarity. For instance, in the case of exposure of aircrew, the source of exposure is ‘existing’ but the action that results in the exposure (the decision to fly) is clearly ‘planned’.

2.5.3. Exclusion, exemption and clearance

The presentations and discussions revealed a general perception among participants that the treatment of exposure to natural sources in international standards was unnecessarily complicated and confusing. This might explain why there seemed to be so many misunderstandings and differences in interpretation of the standards, especially with respect to the concepts of exclusion, exemption and clearance. For instance:

(a) The fact that the table of exempt activity concentration values in the current BSS did not generally apply to NORM was frequently overlooked;
(b) The 1 Bq/g criterion for subjecting material to regulatory consideration as a source within a practice was variously referred to by participants as an exclusion level, an exemption level, a clearance level or even a limit;
(c) There was a tendency to apply the concept of exemption not only to planned exposure situations but to existing exposure situations as well.

From what was said during the presentations and discussion, there appeared to be a particular problem with the use of the terms exclusion and exemption. These terms tended to be used interchangeably and were in any case not really meaningful without addressing the question: ‘exclusion or exemption from what?’ For instance, reference to the 1 Bq/g criterion as an exclusion level could mean either that the material satisfying this criterion was excluded from regulation as a planned exposure situation (but not necessarily from control as an existing exposure situation), or that the material was excluded from the standards entirely. Without appropriate qualification, the term ‘exclusion’ in this context was ambiguous. Similarly, NORM in transport that fell outside the scope of the IAEA Regulations for the Safe Transport of Radioactive Material (the Transport Regulations) was sometimes referred to as being excluded, but such material was not necessarily excluded from the requirements of the BSS.

Interpretation of the concept of clearance was also not without its difficulties. Mention was made of material being cleared from regulatory control, but with conditions applied. This raises the question of whether this is truly clearance, since the imposition of conditions could imply the need for some form of ongoing regulatory control.

2.5.4. The need for an industry specific approach

Another general conclusion was that no single approach to the control of exposure to NORM was appropriate for all industrial processes involved. In the context of education and training it was pointed out that, while grouping various industrial activities under the broad description ‘NORM industry’ may be convenient, it is a significant departure from conventional classification practice. This same argument is valid in a regulatory context, since the nature and level of the radiological risk varies considerably from one industrial process to another. Similarly, it was pointed out on several occasions that actions taken to comply with regulation, under the general banner of ‘good practice’ or ‘best practice’, were situation specific and could not be defined on a more general ‘NORM industry’ basis. There was therefore a strong call for an industry specific approach to the control of exposure to NORM and the ongoing efforts of the IAEA to develop industry specific Safety Reports were acknowledged and welcomed in this regard. A similar approach has been adopted in the Australian Safety Guide on NORM management referred to in Section 2.2, for which separate annexes for various NORM industries are being developed. Two examples highlighted the particular need for an industry specific approach when applying radiation protection measures in challenging operating environments:

(i) The oil and gas industry operates under difficult and diverse environmental conditions in many parts of the world where appropriate regulatory systems are not always sufficiently developed. It was evident that good progress had been made in developing good practices tailored to the industry's own particular set of circumstances.

(ii) Mining operations, particularly those with associated legacy issues, have their own particular challenges, as was evident from presentations on legacy uranium mining sites in Central Europe and a former copper–cobalt mining site in Africa where consideration was being given to a restart of operations.

3. EXPOSURE OF WORKERS

There were many references throughout the symposium to the adoption of a graded approach to the regulation of worker exposures, in line with international standards, and there are now several examples of how this has been implemented in practice. Also, it would appear that the role of general occupational health and safety regulations in controlling radiological hazards at work, particularly in the case of airborne dust control, is becoming increasingly recognized as an important part of the graded approach to regulation. However, the acquisition of exposure data for workers and the assessment of dose still suffer from a non-standardized approach and incomplete information in several countries, making a reliable assessment of the need for, and extent of, regulatory control difficult.

The results of various dose assessments for workers reported in the symposium are summarized in Table 1. The annual effective doses given in Table 1 include the contributions from external exposure to gamma radiation and internal exposure to inhaled dust but (except where stated otherwise) not the contributions from the inhalation of radon and thoron, which are usually treated separately. The magnitudes of the doses are very much in line with those reported elsewhere, including those reported in IAEA Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials, as well as those reported at the NORM V symposium in 2007.

Of particular significance was an investigation carried out in France, covering more than 400 workplaces in seven types of industrial activity. It was found that a third of the annual doses were below 0.1 mSv and half were below 0.25 mSv, while only 15% of the doses were above 1 mSv. It is becoming increasingly clear that, in the majority of industrial workplaces involving NORM, the annual effective dose received by a worker is likely to be less than 1 mSv. In some cases, doses of a few millisieverts per year may be received, but only in relatively rare situations, such as might occur in the uranium, thorium and

TABLE 1. DOSES RECEIVED BY WORKERS, AS REPORTED AT THE SYMPOSIUM

(Excluding doses from inhalation of radon and thoron, except where stated otherwise)

	Annual effective dose (mSv)			
	Minimum	Mean	Maximum	Distribution
Uranium ore mining		7.8	40–45	
Processing of thorium concentrate[a]	3.0		7.8	
Production of thorium compounds[b]			82	67% < 1
Mining of rare earth ore[c]		0.24–1		
Beneficiation of rare earth ore[c]		0.28–0.61		
Handling of monazite			0.3	
Rare earth separation and purification			0.3	
Decommissioning of a rare earths plant[d]	0.2	7.2	8.94	
Mining of ore other than uranium ore			<1	
Oil and gas production, offshore			0.5	
Oil and gas production, onshore			0.05	
Oil production, cleaning of pipes[c,e]		0.6	3	80% < 1
Titanium dioxide pigment production			<0.5	
Titanium dioxide pigment production			0.27	
Phoshate ore storage			0.28	
Phosphate fertilizer production			0.5	
Phosphate industry facilities[f]	0.009		2.7	
Zircon production			0.4	
Bastnäsite (zirconia) production			0.4	
Manufacture and use of zircon and zirconia	0		2.3	87% < 1
Manufacture and use of refractory ceramics	~0.01		1.5	98% < 1
Manufacture of zircon/zirconia ceramics	· · · · · · · · · · · · · · · ·Negligible· · · · · · · · · · · · · · ·			
Processing of Sn, Al, Ti and Nb ores	0		3.2[g]	69% < 1
Copper smelting			<1	
Recycling of metal scrap	· · · · · · · · · · · · · · · ·Negligible· · · · · · · · · · · · · · ·			
Combustion of coal	0		0.4	
Combustion of coal			Generally <1	
Combustion of coal			0.13	
Drinking water treatment			<1	

TABLE 1. DOSES RECEIVED BY WORKERS, AS REPORTED AT THE SYMPOSIUM (cont.)
(Excluding doses from inhalation of radon and thoron, except where stated otherwise)

	Annual effective dose (mSv)			
	Minimum	Mean	Maximum	Distribution
Effluent water treatment, former U mine			<1	
Geothermal water (health spa)			<1	
Manufacture of mineral insulation material[h]	0.0011		0.0173	

[a] Doses include contributions from inhalation of thoron.

[b] Doses >1 mSv/a, mainly due to dust inhalation, were identified in two of the six workplaces investigated. The assessment is being repeated after the implementation of dose reduction measures (equipping workers with respiratory protection, cleaning the workplaces periodically and installing air filtration).

[c] Doses from external exposure only.

[d] Doses received over a 9 month decommissioning period.

[e] Doses received over a 5 month refurbishment period.

[f] Doses include contributions from inhalation of radon.

[g] The maximum dose was 6 mSv prior to 2008.

[h] The minerals were coal, bauxite, basalt and cement.

monazite based rare earths industries, would the dose be likely to exceed 5 or 6 mSv in a year. It was pointed out that this distribution of doses is broadly the same as that observed for work with artificial sources, suggesting that the familiar protection philosophy of engineering controls, working procedures and personal protective equipment (in that order) is equally appropriate for exposure to NORM.

Relatively little new information on radon in workplaces emerged from the symposium. In some instances, doses received by workers from the inhalation of radon were reported separately, from which it can be concluded that the radon concentrations in most of the workplaces concerned (covering the production of zircon, zirconia and refractory ceramics and the processing of ores of metals other than uranium) were generally less than about 100 Bq/m^3. The only workplaces identified as having significantly higher radon exposures were, as might have been expected, uranium mines (in this case uranium mines in the Czech Republic). Analysis of the dose data presented suggests that those mines generally have radon concentrations of up to about 800 Bq/m^3 and very occasionally up to about 2000 Bq/m^3. The mean radon concentration would appear to be around 500 Bq/m^3.

4. EXPOSURE OF MEMBERS OF THE PUBLIC

The results of various estimates of doses received by members of the public (excluding doses from radon) from mining and mineral processing facilities, as well as from the use of residues from such facilities, are summarized in Table 2. As is inevitably the case when assessing doses to members of the public, the estimates are likely to be subject to considerable uncertainty and tend to be conservative, owing to the necessity for modelling of the relevant exposure scenarios. The estimated doses in Table 2 are generally below 1 mSv per year. However, for the use of phosphogypsum in house construction, it is clear that the dose depends strongly on the way in which it is used (as well as on the activity concentration in the phosphogypsum). If it is used in the form of thick, solid building elements, the estimated dose could exceed 1 mSv while, if the phosphogypsum is used more sparingly in the form of thin building elements, the estimated annual dose is well below 1 mSv.

Some data on exposure to radon were also presented. For a house constructed using phosphogypsum panels, it could be inferred from the data that the incremental indoor radon concentration (that is, the contribution from the panels) was likely to be in the range 2–60 Bq/m^3 (the exact value depending on the activity concentration in the phosphogypsum), whereas for a house constructed from more conventional building materials, the contribution from those materials was 0–18 Bq/m^3. It was reported from Australia that earlier studies on the use of phosphogypsum plasterboard had led to the conclusion that the increase in indoor radon concentration was insignificant.

5. TRANSPORT OF NORM AND RADIOLOGICAL SCREENING OF CONSIGNMENTS

The symposium provided participants with the opportunity to learn about the results of an IAEA coordinated research programme on the transport of NORM by way of a general overview paper complemented by individual reports from three of the participating countries (Canada, France and Romania). There was considerable diversity among the studies carried out in the participating countries and this, together with the inevitable uncertainties introduced through the extensive use of exposure modelling, made it difficult to make direct comparisons of the results. However, interim conclusions suggested that the present criterion for application of the IAEA Transport Regulations to NORM (namely, 10 times the activity concentration for exempt material) was generally valid. Some of the results showed that this criterion was unnecessarily strict for at least some of the transport situations involving NORM, implying the need for

TABLE 2. DOSES RECEIVED BY MEMBERS OF THE PUBLIC, AS REPORTED AT THE SYMPOSIUM
(Excluding doses from inhalation of radon)

	Annual effective dose (mSv)
Mining of rare earth ore	0.044
Beneficiation of rare earth ore	0.043
Use of slag from rare earths and steel production in house bricks	~0.2
Production of Th welding rods	Negligible
Mining of ore other than uranium ore	Specified only as <1
Large mineral residue deposit, 1 Bq/g ^{238}U and/or ^{232}Th	0.05–0.26
Oil and gas production	Specified only as <1
Elemental phosphorus production	<0.04
Use of dicalcium phosphate animal feed	<0.02
Use of phosphogypsum for agriculture	Negligible
Use of phosphogypsum (PG) for construction of houses:	
Walls and ceilings, PG panels,	0.02–0.2
Walls, ceilings and floor, hollow PG panels	0.46
Walls, ceilings and floor, solid PG panels	4.5
Walls, PG plasterboard lining	0.15
Walls, PG plasterboard lining	Insignificant
Walls, PG in bricks and cement	≤1.4
Manufacture of zircon/zirconia ceramics	Negligible
Steel production	<0.01
Use of metal recycling slag for road construction	Specified only as <1
Combustion of coal	Negligible
Drinking water treatment	Negligible
Disposal of water treatment residue in landfill	0.01
Effluent water treatment, former U mine	Specified only as <1
Use of common building materials for house construction	<0.3–1

consideration on a case by case basis. Such a provision does not currently exist in the Transport Regulations.

Attention was drawn to the fact that container shipments were now being monitored for radioactive material at major ports around the world, to counter the possible trafficking of illicit nuclear material. Such a monitoring system at the port of Antwerp was described, revealing that this had led to a large number of alarms being triggered due to the presence of moderately radioactive NORM (such as zircon) in the container. A methodology had been developed for determining the activity and activity concentration of each radionuclide of natural origin. It was concluded that this is an area of growing concern worldwide and that continued improvements will be needed in the design and operation of such monitoring systems and in the training of operators.

Similar portal monitoring systems are installed for the screening of commercial vehicles at the entrances to metal scrap recycling facilities. In this case, the objective is to detect the presence of radioactive sources in order to prevent them from becoming inadvertently melted with the scrap. As a follow-up to a presentation on the so-called ‘Spanish Protocol’ made at the NORM V symposium in 2007, a paper was presented on the most recent experience gained in the implementation of this system since its introduction some 10 years ago. The presence of NORM contamination accounted for more than half of all the alarms triggered. The main objective was to be able to determine the activity concentrations of the NORM contaminated items to enable them to be segregated into items that could be recycled by melting and those that had to be removed and transported to a radioactive waste facility. The criterion for segregation was an activity concentration of 1 Bq/g for ^{226}Ra and ^{232}Th. The risk of inadvertent melting of radioactive sources is a worldwide problem and it was clear that the Spanish Protocol approach was a promising way forward in dealing with it on an international scale.

6. USE OF NORM RESIDUES

The use of NORM residues as recoverable resources (co-products), either directly or following some form of treatment or recycling process, was referred to in almost 20 of the 80 papers and poster presentations. It was clear from these references that the concept of using NORM residues rather than disposing of them as waste is gaining increasing acceptance around the world. Much of this was being driven by sustainability issues such as concerns over the depletion of resources, by a growing recognition that the amounts of NORM waste needed to be minimized in order to make their disposal manageable, and sometimes by sheer economic considerations (some of which only become evident once the true costs and liabilities of waste disposal are taken into account). Some countries are now specifically providing for NORM residue recycling and use in their regulatory systems. For instance, it was reported that recent legislation in the Netherlands identifies the use of NORM residues as the primary target of a NORM residue management system. For application in civil engineering, a specific requirement in the Dutch legislation is that the NORM residue is diluted to a level such that it is no longer considered radioactive (in that it does not exceed the relevant ‘exemption’ level). Thus, dilution in this case is not only a treatment option but also a legal obligation. Only if the options of use or recycling are not feasible can the material be disposed of and only then is it considered as waste.

Many instances of NORM residue recycling and use were mentioned in the symposium and are summarized in Table 3.

To ensure that the doses received from the use of the NORM residues remains within acceptable levels, various conditions are being applied or are being considered by the regulatory authorities concerned. The following examples were reported:

(a) In Sweden, historical NORM residues may be used in landfill and construction applications if the activity concentrations of radionuclides in the ^{238}U decay series do not exceed 3 Bq/g, this being 3 times the ‘exemption’ level.
(b) In terms of new regulations in India, the use of phosphogypsum in building materials is permitted if the ^{226}Ra concentration does not exceed 1 Bq/g (after dilution with lower activity material if necessary). Phosphogypsum panels are permitted for house construction if the surface activity concentration does not exceed 40 000 Bq/m^2.
(c) In Spain, contaminated scrap metal may be recycled by melting if the ^{226}Ra and ^{232}Th activity concentrations do not exceed 1 Bq/g.
(d) In the current draft of the proposed new European Council Directive, building materials can be used without restriction if the dose from indoor external exposure does not exceed the background outdoor external exposure by more than 1 mSv per year. If this value is exceeded, control measures should be considered, ranging from registration and general application of relevant building codes to specific restrictions on the use of the material.

The presentations and discussion at the symposium highlighted the placing of restrictions on the use of NORM residues in building materials as an issue that is attracting ever more attention. While there seemed to be a degree of agreement on the value of 1 mSv as a general reference level for building materials, there was less of a common view on how this should be translated into measurable quantities such as activity concentration. A more conservative approach was discernible in European countries compared with some countries elsewhere and even within Europe there were differences. For instance, some countries in Europe are applying, or are considering applying, an additional criterion for

TABLE 3. EXAMPLES OF THE USE OF NORM RESIDUES, AS REPORTED AT THE SYMPOSIUM BY VARIOUS COUNTRIES

	NORM residue	Use
Australia	Coal ash	Landfill, roads, building material
	Zinc smelter slag	Abrasives (discontinued)
	Red mud from bauxite processing	Soil conditioner, landfill
Brazil	Phosphogypsum	Soil amendment, building material
China	Waste rock from rare earths mine	Roads, tailings embankments
	Effluent from rare earths extraction	Recovery of sulphuric acid
	Blast furnace slag	Recovery of iron content, iron-free residue used in cement, bricks and fill after diluting with low-activity residue such as flyash
Germany	Steel slag	Certain types of concrete
	Metal recycling slag	Road construction
	Copper slag	Building material (discontinued)
	Water treatment sludge	Bricks and concrete (1–2%)
	Red mud from bauxite procesing	Bricks (discontinued)
India	Phosphogypsum	Cement (2–5%), plasterboard, fibre-reinforced building panels, plaster of Paris
Netherlands	Zircon sand	Reuse (found not to be viable because of strict quality requirements)
	Various residues	Low quality concrete for basic uses such as separation blocks in the storage of bulk material (under consideration)
Spain	Contaminated scrap metal	Recycled by melting
	Ferrous sulphate monohydrate and heptahydrate from TiO_2 production	Fertilizer, soil amendment, animal feed, flocculant for water treatment
	Red gypsum from TiO_2 production	Setting retardant in cement
Sweden	Historic residues: burnt alum shale, mining and steel residues, phosphogypsum, lightweight concrete based on alum shale, material from bedrock drilling	Landfill, construction materials

building materials to specifically control radon exposure. It was pointed out that, as a result of new recommendations by the ICRP and the World Health Organization, reference levels for indoor radon are undergoing a downward trend. This has heightened concerns within some European countries that a restriction based only on external exposure might not be sufficient to adequately control radon exposure. There also appeared to be different views on whether the 1 mSv dose criterion should refer to the total external dose from the building material or just the contribution from NORM contained within it.

Several presentations described work being undertaken to assess the implications of using phosphogypsum for agricultural and building construction purposes. In the case of agricultural use as a soil amendment, various experiments on soils and plants had been carried out. The results of these experiments suggested that radionuclides such as ^{226}Ra, ^{228}Ra and ^{210}Pb introduced into the soil via the addition of phosphogypsum exhibited low mobility and had a low availability for uptake by plants. Measurements conducted on plants grown in the amended soils showed no significant increase in radionuclide concentrations. In the case of building construction applications, various dose assessments were reported, the results of which are included in Table 2.

7. MANAGEMENT OF NORM RESIDUES DESIGNATED AS WASTE

Various presentations referred to the treatment, storage and disposal of NORM residues for which recycling and use was not a feasible option and which were therefore designated as waste. Many such residues existed as legacy situations from former industrial activities. The situation in Central Asia regarding former uranium production sites was highlighted as a major challenge in this regard, requiring coordinated international effort to assist the countries concerned in planning and carrying out the necessary remediation work. With regard to the establishment of good practices for the management of NORM waste, it was emphasized on several occasions that a risk based approach to the disposal of NORM waste was essential, that non-radiological hazards nearly always had to be taken into account and that a situation specific approach had to be adopted, even though the general principles and safety standards involved were common to all situations,.

It was interesting to note that, for the symposium as a whole, considerably more attention was given to the recycling and use of residues than to their disposal as waste. This appears to be the first time that this has happened in this series of symposia and reflects an important shift in philosophy away from the more traditional approach in which most NORM residues were automatically looked upon as waste.

Several types of NORM wastes were mentioned in the presentations, including:

(a) Tailings and other waste from the processing of uranium ore;
(b) Tailings, slag and chemical processing wastes associated with the production of thorium and rare earths;
(c) Radium-rich scale from the oil and gas industry;
(d) Sludge from water treatment facilities.

A reasonably clear picture emerged from the symposium regarding the most commonly used (and accepted) options for disposal of NORM waste, which can be summarized as follows:

(a) For large volumes of relatively low activity waste, such as mine tailings, the only two practicable options available were for it to be isolated in above ground, custom built containments such as tailings dams or to be diluted with non-radioactive soil or sand and returned into the remediated land form. The latter option is accepted practice for mineral sand tailings.
(b) Low and intermediate volumes of relatively high activity NORM waste such as pipe scale from the oil and gas industry and process residue from the extraction of rare earths and thorium were usually disposed of in one of three ways:
    (i) By emplacement in underground radioactive waste repositories such as that described in a presentation from Norway;
    (ii) By emplacement in shallow ground, engineered (usually concrete) structures such as those described in a paper from India.
    (iii) In the case of pipe scale from the oil and gas industry, by reinjection into the formation using a process known as ‘slurry fracture injection’.
(c) Moderate volumes of NORM waste with low activity concentrations (but above the applicable exemption or clearance level) were increasingly being authorized for disposal in conventional disposal facilities for industrial or hazardous waste, such as landfill sites, sometimes with some additional, relatively simple protection measures being applied to cater for the radionuclide content. In all cases reported, the upper bound on the radionuclide activity concentration was being set at 10 times the exemption or clearance level (the actual or proposed value of which varied between countries — 1 Bq/g in Sweden and the Netherlands and 0.5 Bq/g in Norway). Thus the actual or proposed upper bound on activity concentration for this form of disposal was either 5 or 10 Bq/g.

8. MAIN FINDINGS OF THE SYMPOSIUM

8.1. Regulatory aspects

(a) Further progress has been made towards the harmonization of standards and regulatory approaches for the control of exposure to NORM, but this progress is by no means universal and there is still a long way to go.

(b) The revision of international standards is seen as an opportunity for establishing a more substantial platform on which to base national standards and regulations on NORM. The hope among the symposium participants was that the revised standards would provide greater clarity and detail regarding NORM, but there are concerns that the standards will still suffer from being too complicated and open to misinterpretation. There have been difficulties in attempting to take account of the new (2007) recommendations of the ICRP in the revision process, particularly with regard to distinguishing between planned exposure situations and existing exposure situations. In this regard, the formation of an ICRP Task Group on NORM, while seen as long overdue, was nevertheless welcomed by the symposium participants.

(c) A listing of industrial processes involving NORM that are most likely to need regulation as practices, first put forward by the IAEA in 2006, has now received widespread acceptance, thus providing national authorities with the means to focus their regulatory attention on those areas where it is most needed. In particular, there is a growing acceptance of the 1 Bq/g criterion for uranium and thorium series radionuclides as a tool for determining which industrial process materials need to be considered for regulation. However, concerns were expressed that this progress could be undermined by those continuing to advocate an over-ly cautious approach based on questionable risk assessments derived from conservative modelling and implausible exposure scenarios.

(d) There is growing recognition that an industry driven (or even process driven) approach is needed for ensuring that exposures to NORM are controlled sensibly and effectively. Industrial activities involving NORM, and legacy situations from such activities, are very diverse and each has to be addressed by developing ‘good practice’ according to the particular set of circumstances — there is no such thing as universal best practice for NORM.

8.2. The expanding knowledge base on exposures in NORM industries

Assessments of worker and public doses arising from the processing of minerals and raw materials continue to be made, although there are still some deficiencies and discrepancies in the way in which data are gathered. Some of the assessments of worker doses continue to be compromised by reliance on exposure modelling rather than on representative measurements in the workplace. The results of the assessments, which confirm previous findings, can be summarized as follows:

(a) In the majority of situations, the annual doses received by workers are below 1 mSv, with doses of a few millisieverts being received in some cases. Only in relatively rare situations, such as may occur in the uranium, thorium and monazite based rare earths industries, would there be a significant possibility of doses exceeding 5 or 6 mSv per year.

(b) The data on exposure to radon in workplaces involving NORM suggest that, with the exception of uranium mines, radon is not a significant source of exposure, with activity concentrations generally below 100 Bq/m^3.

(c) Assessments of doses received by members of the public continue to suggest that annual doses are consistently and significantly below 1 mSv. Many of the results were reported as ‘negligible’ or ‘insignificant’.

8.3. Transport of NORM

(a) As a result of an IAEA coordinated research programme on the transport of NORM, there is now a body of knowledge available that provides some scientific evidence to support the criterion defining the scope of application of the Transport Regulations to NORM (namely, 10 times the activity concentration for exempt material).

(b) Transport of NORM is falling increasingly under the spotlight because of the triggering of alarms designed to detect radioactive sources in scrap metal or to combat the trafficking of illicit nuclear material. The scrap management system developed in Spain (the ‘Spanish Protocol’) is setting an example for how this problem can be addressed on an international scale. Detection systems are becoming increasingly widespread and sophisticated and progress is being made in developing techniques for identifying, characterizing and allowing the passage of NORM without compromising the effectiveness of the detection systems.

8.4. Use of NORM residues

(a) There is increasing recognition of the need to regard NORM residues as a resource rather than as waste. For the first time in this series of symposia, more attention was given to the use of NORM residues than to their disposal as waste and many examples of such use were provided. National legislation is already changing in some countries, moving away from restriction or prohibition of the use of NORM residues towards acceptance and even encouragement of such use (including dilution with lower activity material where necessary).

(b) There is a lack of uniformity in the approach to the use of NORM as a component of building material, although it is generally agreed that any situation giving rise to a dose of more than 1 mSv per year would need special consideration and in certain cases some form of restriction. Information provided at the symposium seems to suggest that the approach to the use of NORM in building materials, while in all cases taking radiological considerations into account, tends to be more conservative in countries with highly developed economies and more realistic and pragmatic in countries with emerging economies.

(c) The use of phosphogypsum as a co-product of phosphoric acid production continues to be investigated in some detail. The information presented at the symposium confirms earlier findings in that the use of phosphogypsum in agriculture can be of great benefit for crop production while having no significant radiological impact. A considerable amount of new information is now available on the use of phosphogypsum as a building material or as a component of such material. The findings suggest that, although the activity concentration of the phosphogypsum is an important factor, the way in which it is used has a strong influence on the incremental exposure level in the building concerned, with doses ranging from ‘insignificant’ to more than 1 mSv per year. On the strength of this information, one country has recently established specific criteria for permitting the use of phosphogypsum in building materials, so as to maintain radiological safety without losing the considerable social and economic benefits to be gained from such use.

8.5. Disposal of NORM residues as waste

(a) A reasonably clear picture is now emerging on the options available for disposal of NORM residues as waste.

(b) The choice of disposal option is often specific to a particular industry. For instance, the oil and gas industry makes use of ‘slurry fracture injection’ into the geological formation to dispose of high activity pipe scale, while the mineral sands industry dilutes its mineral processing tailings with low activity sand or soil and returns it to the mining void.

(c) Increasing use is being made of disposal in conventional landfill facilities established for industrial or hazardous waste, sometimes with some additional radiation protection measures being applied. Acceptance criteria for landfill disposal, expressed in terms of maximum radionuclide activity concentration, have been established in several countries, with values ranging from 5 to10 Bq/g.

OPENING SESSION

Chairpersons

J. HILTON
United Kingdom

K. EL MEDIOURI
Morocco

OPENING ADDRESS

M. Loudiki
Faculty of Sciences Semlalia, Cadi Ayyad University,
Marrakesh, Morocco

On behalf of the president of the University, who apologizes for not being here today, I am delighted to welcome you to Marrakesh where you have decided to hold this 6th International Symposium on Naturally Occurring Radioactive Material, NORM VI.

On behalf of Cadi Ayyad University, I would like to thank the symposium organizers, Hassan II University of Mohammadia, CNESTEN, CERPHOS, OCP and the IAEA and all the sponsors who have made it possible for this important scientific event to take place in Marrakesh, the home of our institution.

The Faculty of Sciences, Semlalia, has a great interest in training and scientific research in the radioactive and nuclear material domain. Our academic programme includes various degrees with several courses in this discipline and our laboratories are also conducting advanced research in this area.

This international meeting allows, no doubt, the researchers, industry experts and professionals to expose their ideas and scientific results concerning new methods and technologies for the extraction and processing industry involved with naturally occurring radioactive material.

The items in the symposium programme are also important to universities, research centres, industrial enterprises and governmental and non-governmental institutions. This will certainly be an opportunity to reflect on a framework of partnership and cooperation in order to work together for developing new projects and to ensure their implementation.

On behalf of the Faculty of Sciences, Semlalia, I wish you all success. I also hope that this symposium will allow further success to be achieved in future symposia, as well as the perpetuity of scientific and technologic exchange and the reinforcement of ties between the participants and all the countries represented here.

To conclude I would like to thank again the symposium organizers, especially M. Ali Misdaq and all the members of his research team. I congratulate them and wish them great success. I wish you a good stay in Marrakesh.

Thank you very much.

OPENING ADDRESS

A. Fahli
Hassan II University,
Mohammedia, Morocco

I speak on behalf of the President, Ms Rahma Bourqia, who apologizes for not being with us this morning. In her absence, I was given the task of conveying to you her greetings, her welcome to you in Morocco and her wishes for success during this crucial NORM VI symposium.

I am not able to embark on the explanation, outcomes and beneficial effects of the topics on NORM. It is for you, the specialists, to debate that in depth during the next four days.

We would like to thank all the experts and prominent researchers who responded to our invitation and came from faraway places around the world. I wish them a pleasant stay in Morocco and especially in Marrakesh.

Our country is particularly interested in this subject. It is witnessing several meetings aimed at debating the National Charter of the Environment and for Human Development, initiated and launched by our king, His Majesty Mohammed VI. Moreover, this subject also interests our research teams at various laboratories. I hope this occasion will be seized upon to identify areas of collaboration and to establish partnerships.

On this occasion, I would like to thank all those who have helped us to achieve the organization of this major scientific event, especially the Steering Committee members who were closely involved in the organization process. They provided real support to us through their guidance and suggestions, so let me thank them once again.

This event benefited from the moral support and sometimes material support of various institutions, Ministries and NGOs having an association with the topic of the symposium. I wish to thank in particular the IAEA; the Ministry of Higher Education, Scientific Research and Staff Training; the Hassan II Academy of Sciences and Techniques; the CNRST; and the academic associations AIGAM, AMR, AMPST and GASUP. I would also like to thank our official sponsor, the OCP Group, whose support was crucial for us. Finally, I would like to thank the members of the National Organizing Committee, the administrative staff of the two universities and all others who contributed, each from their own locations, to the success of this meeting.

Thank you very much indeed for your attention.

OPENING ADDRESS

Y. Bouabdellaoui
Institute Agronomique et Vétérinaire Hassan II,
Rabat, Morocco

On behalf of the National Organizing Committee, good morning and thanks a lot for coming. The convening of this conference in Marrakesh constitutes, without doubt, a further step towards the promotion of scientific research and development in the field of NORM activities as well as furthering the aim of improving their radiation safety aspects. It is obvious that the main objective of this gathering is how to strengthen and improve protection of humans and the environment and likewise to ensure sustainable development through the utilization of NORM.

Ladies and gentlemen, our country Morocco is launching many action plans for economic and social development. Therefore, the convening of this Conference responds to this strategic interest. Morocco is committed to the achievement of comprehensive and sustainable development of the legal and regulatory framework for the protection of humans and the environment.

Ladies and gentlemen, as stated by the organizers — the Steering Committee and the National Organizing Committee — and given the findings of NORM V held in Seville, the objectives of this NORM VI symposium are to provide a forum for debate between the scientific, technical and regulatory communities involved in NORM issues. It gives the opportunity to present and discuss recent studies and work achieved, to share results and experience and to identify future needs and challenges. With your involvement, we expect that this symposium, like previous symposia in this series, will lead to increased harmonization of regulatory approaches and the wider application of international standards, especially with regard to discussion of the draft version of the revised BSS (IAEA Safety Series No. 115) as proposed by the IAEA and the other cosponsors.

I would also like to take this opportunity, ladies and gentlemen, to express my appreciation to the Steering Committee and the Scientific Committee for their involvement and their valuable help and expertise in preparing for this event and to the IAEA for their assistance and cooperation in contributing to the success of this meeting.

But let me tell you that the main contribution to this success comes from your attendance and your high standard of participation; So our thanks go as well to all the participants who came from different and remote parts of the world, extending to you a warm welcome and wishing you a pleasant stay in our country and in the fascinating city of Marrakesh. My thanks go also to our partner and official sponsor, the OCP group.

Thank you, ladies and gentlemen.

KEYNOTE ADDRESS

REVISION OF THE INTERNATIONAL BASIC SAFETY STANDARDS AND IMPLICATIONS FOR EXPOSURE TO NATURAL SOURCES

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Abstract

The International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS) were published by the IAEA in 1996. Following a review in 2005–2006, a decision was made to embark on a revision process starting in 2007 in collaboration with the co-sponsoring organizations. Completion of a final draft (in readiness for approval by the IAEA Board of Governors) is expected in 2011. The requirements in the new BSS draft are in line with the 2007 Recommendations of the ICRP. The three types of exposure situation recommended by the ICRP — planned exposure situations, existing exposure situations (currently referred to as chronic exposure situations) and emergency exposure situations — are reflected in the structure of the document. Exposure to natural sources continues to be generally subject to the requirements for *existing exposure situations* but exposure control, rather than being based on the use of action levels as at present, is based instead on the use of reference levels (defined by the ICRP as levels of dose or risk above which it is judged inappropriate to allow exposures to occur and below which optimization of protection should be implemented). The maximum reference levels for exposure to radon are expressed in terms of radon activity concentration and are set at 300 Bq/m^3 for homes and 1000 Bq/m^3 for workplaces, these values corresponding to an effective dose of about 10 mSv per year. For exposure to radionuclides in commodities, a maximum reference level of about 1 mSv per year is applicable, ensuring a level of protection similar to that for planned exposure situations, even though the mechanism of control is different. Some basic requirements for cosmic ray exposure of aircrew and space crew are included for the first time. The following exposures are, by exception, subject to the requirements for *planned exposure situations*: public exposure to radioactive discharges and waste; occupational exposure to radon when its concentration exceeds 1000 Bq/m^3 or when required by or directly related to the work; and exposure to material (other than commodities such as food, drinking water, fertilizers and construction materials) with a radionuclide activity concentration exceeding 1 Bq/g (or 10 Bq/g in the case of ^{40}K). For the first time, numerical criteria for exemption and clearance of NORM have been included. Exemption is determined on the basis of dose commensurate with natural background levels (about 1 mSv per year). Clearance criteria for NORM (first published in 2004 in Safety Guide No. RS-G-1.7) are 1 Bq/g for U and Th series radionuclides and 10 Bq/g for ^{40}K. It is concluded that the revised BSS should provide greater clarity on the control of exposure to natural sources and, as a result of the new 'reference level' approach, the level of protection in existing exposure situations such as indoor radon will be significantly increased.

1. INTRODUCTION

1.1. The International Atomic Energy Agency (IAEA)

The IAEA is an independent, intergovernmental, science and technology based organization that serves as the global focal point for nuclear cooperation. It currently has 151 Member States and employs 2326 staff. The IAEA was set up as the world's 'Atoms for Peace' organization in 1957 within the United Nations family. The IAEA works with its Member States and multiple partners worldwide to promote safe, secure and peaceful applications of nuclear technologies. The IAEA Statute, the original version of which was approved by 81 nations in 1956, outlines the three pillars of the IAEA's work:[1]

(a) Safeguards and verification: The IAEA is the world's nuclear inspectorate, with more than four decades of verification experience. Inspectors work to verify that safeguarded nuclear material and activities are not used for military purposes.

(b) Safety and security: The IAEA helps countries to upgrade nuclear safety and security, and to prepare for and respond to emergencies. Work is keyed to international conventions, standards and expert guidance. The main aim is to protect people and the environment from harmful radiation exposure. In the safety area, the IAEA's activities cover nuclear installations, radioactive sources, radioactive materials in transport, and radioactive waste. A core element is establishing international safety standards for the management and regulation of activities involving nuclear and radioactive materials and promoting the application of these standards.[2]

(c) Science and technology: The IAEA helps countries to mobilize peaceful applications of nuclear science and technology. The work contributes to goals of sustainable development in the fields of energy, environment, health and agriculture, among others, and to cooperation in key areas of

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[1] More information can be found on the IAEA's website http://www.iaea.org.

[2] The IAEA is authorized in terms of its Statute to "establish or adopt, in consultation and, where appropriate, in collaboration with the competent organs of the United Nations and with the specialized agencies concerned, standards of safety for protection of health and minimization of danger to life and property (including such standards for labour conditions), and to provide for the application of these standards to its own operations as well as to the operations making use of materials, services, equipment, facilities, and information made available by the IAEA or at its request or under its control or supervision; and to provide for the application of these standards, at the request of the parties, to operations under any bilateral or multilateral arrangement, or, at the request of a State, to any of that State's activities in the field of atomic energy".

nuclear science and technology. The main areas of activity are technical cooperation, research and development and electricity generation. Through its technical cooperation activities, the IAEA supports cooperative projects achieving tangible social and economic benefits for people in developing countries. Many channels and partnerships provide expert services, specialized equipment, training and other types of support.

The IAEA's relationship with the UN is regulated by special agreements. In terms of its Statute, the IAEA reports annually to the UN General Assembly and, when appropriate, to the Security Council regarding non-compliance by States with their safeguards obligations as well as on matters relating to international peace and security.

1.2. Safety standards established by the IAEA

The safety standards established by the IAEA provide support for Member States in meeting their obligations under general principles of international law. These standards also promote and ensure confidence in safety and facilitate international commerce and trade. The standards reflect an international consensus on what constitutes a high level of safety for protecting people and the environment from harmful effects of ionizing radiation. They are issued in the IAEA Safety Standards Series, which has three categories:

(1) Safety Fundamentals: These present the fundamental safety objective and principles of protection and safety, and provide the basis for the Safety Requirements;
(2) Safety Requirements: An integrated and consistent set of Safety Requirements establishes the requirements that must be met to ensure protection of people and the environment, both now and in the future;
(3) Safety Guides: These provide recommendations and guidance on how to comply with the Safety Requirements, indicating an international consensus that is necessary to take the measures recommended (or equivalent alternative measures). The Safety Guides present international good practices, and increasingly they reflect best practices, to help users striving to achieve high levels of safety.

Supporting publications on protection and safety are issued in other series, in particular the IAEA Safety Reports Series. While not forming part of the IAEA Safety Standards, Safety Reports may describe good practices and give practical examples and detailed methods that can be used to meet safety requirements.

1.3. The International Basic Safety Standards

The International Basic Safety Standards (BSS) have been developed from widely accepted radiation protection and safety principles, especially those established through the recommendations of the International Commission on Radiological Protection (ICRP) and through other IAEA Safety Standards. The BSS are intended to ensure the safety of all types of radiation sources and, in doing so, to complement more specific standards developed for large and complex radiation sources, such as nuclear reactors and radioactive waste management facilities.

The Board of Governors of the IAEA first approved basic safety standards in June 1962; they were published by the IAEA as Safety Series No. 9 [1]. Revised versions were published in 1967 and 1982. In 1996, these standards were superseded by the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, which were jointly sponsored by the Food and Agriculture Organization of the United Nations, the IAEA, the International Labour Organization, the Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, the Pan American Health Organization and the World Health Organization, and published by the IAEA as Safety Series No. 115 [2]. Although Safety Series No. 115 was published before the establishment of the categorization scheme reflected in the current IAEA Safety Standards Series, it is to all intents and purposes a Safety Requirements publication.

2. DEVELOPMENT OF THE REVISED INTERNATIONAL BASIC SAFETY STANDARDS

2.1. Revision process

The IAEA reviews safety standards periodically to assess the need for their revision. This is accomplished through Member State feedback and various IAEA safety committees. In 2005 and 2006, a review of Safety Series No. 115 was made to determine whether there was a need for revision. It was concluded that there was no single urgent reason for change, but the need for several updates and improvements was identified. In particular, there was a need to take account of the newly-published IAEA Fundamental Safety Principles [3] and the anticipated new ICRP recommendations for a system of radiological protection — later to be published as the 2007 Recommendations [4]. There was also a need to incorporate the BSS into the current IAEA Safety Standards Series (as a Safety Requirements document). A revision plan was formulated in November 2006 and the revision process itself started in 2007. The process was coordinated by the IAEA with the full involvement of the organizations that had co-sponsored Safety Series No. 115 and two potential new co-sponsors, the European Commission and the United Nations Environment Programme.

Some guidelines for the revision process were established:

(a) The BSS should continue to provide the international benchmark for radiation safety standards across all circumstances of exposure to radiation and should continue to be written in a regulatory format. In other words, these standards should retain their stand-alone character and be suitable for direct incorporation into national legislation and regulations.
(b) The need for stability in international standards should be recognized, since changes to national laws and regulations take several years to accomplish. Consequently, substantive changes to the BSS should be made only where fully justified in terms of necessary improvements in the level of protection.
(c) The 2007 Recommendations of the ICRP should be followed to the extent possible.
(d) Particular attention should be given to the needs of developing countries.

In order to facilitate the revision process, a BSS secretariat was set up, comprising representatives of the IAEA and the collaborating organizations (the existing co-sponsors and potential new co-sponsors). The BSS Secretariat ensures that the interests, views and responsibilities of each collaborating organization are taken into account, provides a forum for such organizations to inform each other of relevant developments and coordinates the approval process of each collaborating organization. Throughout the revision process, steps have been taken to involve developing countries through various technical meetings and regional workshops.

Drafting groups comprising experts from the IAEA and the collaborating organizations completed a first draft of the revised BSS in mid-2008; this draft was then reviewed by the IAEA safety committees and within the collaborating organizations. More than 1200 written comments were received from 21 countries and 9 organizations. All of these comments were considered during the development of a second draft, which was completed in May 2009. This draft was reviewed in a process similar to that followed for the first draft, and a further 1000 comments were elicited (although many of these were editorial in nature or were repeats of previous comments). Taking these comments into account, a third draft was completed in January 2010. This has been sent formally to all IAEA Member States for comment by the end of May 2010.

Once all comments have been addressed, each co-sponsoring organization will have to complete its own approval process before it is possible to publish the new standards as a Safety Requirements document in the IAEA Safety Standards Series. Within the IAEA, it is intended that the final review of the IAEA safety committees will be complete by the end of 2010, at which point the new BSS will be submitted via the IAEA's Commission on Safety Standards to the IAEA Board of Governors for approval.

2.2. Structure of the new BSS

The structure of Safety Series No. 115 evolved to a large extent from the radiation protection approach advocated by the ICRP in its 1990 recommendations [5]. This approach differentiated between 'practices' and 'intervention'.[3]

The structure of the new BSS draft reflects the radiation protection approach now advocated by the ICRP in its 2007 Recommendations. This approach evolved from the previous process based approach of practices and intervention to one based on the characteristics of radiation exposure situations. In terms of this new approach, the intention of the ICRP was that the implementation of protection for what had previously been categorized as intervention would be enhanced by increasing the attention given to those features common to all situations of exposure. Three situations of exposure are identified: planned exposure situations involving the deliberate introduction and operation of sources; emergency exposure situations that may occur during the operation of a planned situation, from a malicious act or from any other unexpected situation and require urgent action; and existing exposure situations that already exist when a decision on control has to be taken.

The structure of the revised BSS also makes provision for general requirements for protection and safety that apply, regardless of the type of exposure situation. These general requirements include requirements concerning the legal and governmental framework (see Section 2.3.1).

As was the case for Safety Series No. 115, the structure reflects the distinction between occupational exposure, medical exposure and public exposure, a distinction which remains unchanged in both the 1990 and 2007 Recommendations of the ICRP.

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[3] Practices refer to activities that increase the overall exposure to radiation, either by introducing whole new blocks of sources, pathways and individuals, or by modifying the network of pathways from existing sources and thus increasing the exposure of individuals or the number of individuals exposed. Intervention refers to activities that decrease the overall exposure by removing existing sources, modifying pathways or reducing the number of exposed individuals.

The basic structure of the revised BSS can be summarized as follows:

(a) Introduction;
(b) General requirements for protection and safety;
(c) Planned exposure situations:
  - Generic requirements;
  - Occupational exposure;
  - Public exposure;
  - Medical exposure;
  - Emergency exposure situations;
  - Generic requirements;
  - Public exposure;
  - Exposure of emergency workers;
  - Transition from an emergency exposure situation to an existing exposure situation;
  - Existing exposure situations;
  - Generic requirements;
  - Public exposure;
  - Occupational exposure.

2.3. Main changes from Safety Series No. 115

2.3.1. Legal and governmental framework

Safety Series No. 115 contained no requirements on the legal and governmental framework. This framework was addressed only in a Preamble, which did not form part of the requirements. In drafting the new BSS, the scope has been expanded to include some basic requirements in this area. More detailed requirements are contained in another Safety Requirements document that has been submitted to the IAEA Board of Governors for approval [6]. The responsibilities associated with these new requirements can be summarized as follows:

(a) The government is responsible for establishing and maintaining a legal, regulatory and organizational framework for protection against radiation risks and for establishing an independent regulatory body;
(b) The regulatory body is responsible for establishing or adopting regulations and guides for protection and safety and for establishing a system to ensure their implementation;
(c) ‘Users’ (registrants, licensees and others) are responsible for ensuring overall protection and safety, for establishing and implementing a

protection and safety programme and for integrating protection and safety into the overall management system.

2.3.2. *Exemption and clearance*

Numerical criteria for exemption of radioactive material in Safety Series No. 115 addressed only moderate quantities of material and their application to material containing radionuclides of natural origin was very limited. In particular, there were no numerical criteria that were generally applicable to NORM. Requirements on clearance were qualitative only, since no numerical criteria had been established at the time of publication of Safety Series No. 115. In drafting the new BSS, additional numerical criteria for radionuclides of artificial and natural origin have been incorporated, including, for the first time, numerical criteria for clearance. Most of these additional criteria had been established in 2004, but were published only at the Safety Guide level [7].

2.3.3. *Radiation generators and radioactive sources*

New requirements relating to radiation generators and radioactive sources have been added. These new requirements have their origin in the IAEA Code of Conduct on the Safety and Security of Sources, published in 2004 [8].

2.3.4. *Use of radiation for human imaging for non-medical purposes*

The use of radiation for human imaging for purposes other than medical diagnosis or treatment includes its use for occupational, legal or health purposes, for theft detection purposes, or for security or anti-smuggling purposes. Safety Series No. 115 contained one requirement on human imaging for theft detection purposes, which deemed such a practice to be unjustified while at the same time requiring it to be controlled as occupational or public exposure. In drafting the new draft BSS, human imaging for non-medical purposes has been addressed in more detail and the position clarified by stating that, in the case of theft detection, the practice is not justified or, in other situations, is justified only under exceptional circumstances. Non-medical imaging is becoming more widespread (for instance at airport security checks), but such practices are nevertheless still widely viewed as being unjustified. This remains a controversial issue that has proved to be difficult to resolve in the drafting of the new standards.

2.3.5. Existing exposure situations: Action levels and reference levels

The requirements for intervention in Safety Series No. 115 include specific requirements for ‘chronic exposure situations’. These situations are now referred to in the new BSS draft as ‘existing exposure situations’, in line with the terminology used in the 2007 Recommendations of the ICRP. The requirements for chronic exposure situations in Safety Series No. 115 include the concept of ‘action levels’, these being levels of dose rate or activity concentration at or below which remedial action (and thus the need for optimization) was not normally necessary. In the 2007 Recommendations of the ICRP, the concept of action levels is abandoned in favour of ‘reference levels’, which are defined as levels of dose or risk above which it is judged to be inappropriate to plan to allow exposures to occur, and below which optimization of protection should be implemented. Clearly, the concepts of action levels and reference levels are fundamentally different, but there is a tendency for this change to be dismissed simply as a change in terminology because the ICRP, in moving from action levels to reference levels, has essentially maintained the same numerical values of dose (for instance, a maximum of 10 mSv per year in the case of indoor radon exposure). This has resulted in a very significant increase in the stringency of protection measures in existing exposure situations, principally through the removal of what was effectively a lower bound on the application of the optimization process. The new BSS draft follows exactly the same ‘reference level’ approach of the ICRP, with the same values of dose, and therefore reflects the same increase in the stringency of control.

2.3.6. Existing exposure situations: Reference levels for indoor radon

In Safety Series No. 115, guideline numerical values for radon action levels were in the range of 200–600 Bq/m^3 for homes and 1000 Bq/m^3 for workplaces. At the time, these values were assumed to correspond to dose levels of 3–10 mSv per year for homes and 6 mSv per year for workplaces. In drafting the new BSS, these action levels have been replaced by reference levels of 300 Bq/m^3 or less for homes and 1000 Bq/m^3 or less for workplaces. The ICRP, after reviewing recently available scientific information, now considers the dose per unit activity concentration of inhaled radon to be significantly higher than previously assumed, as a result of which it considers these new activity concentration values to be consistent with a dose of around 10 mSv per year [9]. Since reference levels are conceptually different from action levels (see Section 2.3.5), a direct comparison of the old and new activity concentration values is not meaningful, but it is clear that the new requirements will substantially increase the stringency of protection against indoor radon.

2.3.7. Existing exposure situations: Remediation of contaminated areas

Compared with Safety Series No. 115, the new BSS draft contains many more requirements for existing exposure situations. This is to a large extent due to the addition of several new requirements specific to the remediation of areas contaminated by residual radioactive material. These new requirements originate from an IAEA Safety Requirements document published in 2003 [10]. It is intended that, once the new BSS are published, this Safety Requirements document will be superseded.

2.3.8. Existing exposure situations: Occupational exposure to cosmic rays

Occupational exposure to cosmic rays was not covered in Safety Series No. 115. In drafting the new BSS, two requirements have been added, one for exposure of aircrew and the other for exposure of space crew. Owing to the special nature of this type of exposure, it is not appropriate to make very specific requirements in this area. Essentially, the relevant regulatory body is required to determine the need for, and extent of, specific protection measures according to the particular circumstances.

2.3.9. Emergency exposure situations

Requirements in this area have been completely revised and updated. Following a similar approach to that for existing exposure situations, the new text includes requirements on the use of reference levels, in line with the 2007 Recommendations of the ICRP.

2.3.10. Changes to improve user-friendliness

In response to feedback from Member States, changes have been incorporated into the new draft BSS to make the document easier to read and interpret. Requirements that were previously contained in Appendices are now grouped under the relevant headings in the main text. Each requirement is now expressed such that the party responsible for satisfying that requirement is clearly identified. This was not always the case in Safety Series No. 115. In addition, a set of overarching requirements has been established throughout the document, each of which is generally supported by a group of more detailed requirements. The current draft contains 52 such overarching requirements, which are individually numbered and appear in bold text.

3. IMPLICATIONS FOR EXPOSURE TO NATURAL SOURCES

3.1. Exclusion from the scope of the BSS

As in the case of Safety Series No. 115, exposures that are unamenable to control are excluded from the scope of the new BSS draft. While in principle, exclusion can apply to both artificial and natural sources, most practical examples involve natural sources. Examples of excluded exposures are given as exposure to ^{40}K in the body and to cosmic radiation at the surface of the earth. The treatment of exposures that are unamenable to control remains essentially unchanged.

3.2. Planned exposure situation versus existing exposure situation

3.2.1. General approach

For those exposures to natural sources that are not excluded from the scope of application of the new BSS, the general approach is to apply the requirements for *existing exposure situations*. This approach is the same as that adopted in Safety Series No. 115. While there are exceptions to this general approach, these are kept to a minimum in the interests of practicality, as explained in the introduction to the new BSS:

> "……some exposures to natural sources may have characteristics of both planned exposure situations and existing exposure situations. In these Standards, the most appropriate type of exposure situation has been assigned taking practical considerations into account."

The requirements for existing exposure situations apply to exposure to radon in homes and in most workplaces, and to exposure to cosmic rays (other than exposure to cosmic rays at ground level, which is excluded). The requirements for existing exposure situations also apply to exposures to the following materials containing radionuclides of natural origin:

(a) Commodities including food, feed, drinking water, agricultural fertilizer and soil amendments, and construction material, irrespective of the activity concentration;
(b) Residual radioactive material in the environment, irrespective of the activity concentration, from past activities that were not regulated (or not regulated according to current standards);

(c) Any other material, provided that its radionuclide activity concentration does not exceed 1 Bq/g for each radionuclide in the ^{238}U and ^{232}Th series and 10 Bq/g for 40K.

In line with the 2007 Recommendations of the ICRP, the new BSS draft requires that reference levels (see Section 2.3.5) typically have to be set in the range of 1–20 mSv per year. For radon in homes the reference level should not exceed about 10 mSv per year, while for commodities containing radionuclides of natural origin, the reference level should generally not exceed about 1 mSv per year.

3.2.2. Exceptions to the general approach

There are some human activities giving rise to exposure to natural sources that have the characteristics of practices and for which, by exception, control in accordance with the requirements for *planned exposure situations* is the more appropriate and practical approach. In Safety Series No. 115, such exceptions were specified as being human activities that give rise to the following exposures:

(1) Public exposure delivered by effluent discharges or the disposal of radioactive waste arising from a practice involving natural sources;
(2) Occupational exposure to radon required by or directly related to the work;
(3) Occupational exposure to radon incidental to the work, if the exposure is higher than the 1000 Bq/m^3 action level (see Section 2.3.6);
(4) Any other occupational exposure specified by the regulatory body.

In the new BSS draft, items (1)–(3) remain essentially unchanged except for the replacement, in item (3), of ‘action level’ by ‘reference level’ (see Section 2.3.5). However, item (4) is replaced by a new, more quantitative specification that reads as follows:

> “Exposure to material other than food, feed, drinking water, agricultural fertilizer and soil amendments, construction material and existing residues in the environment…………where the activity concentration in the material of any radionuclide in the uranium and thorium decay chains is greater than 1 Bq/g or the activity concentration of ^{40}K is greater than 10 Bq/g.”

This new specification is designed to cover the use of NORM in those industrial applications where the activity concentration of the material involved is above normal levels found in the environment. These applications are typically found in mining and minerals processing, oil and gas extraction and water treatment.

By treating the above-mentioned human activities as practices and subjecting them to the requirements for planned exposure situations, they are required to be controlled according to the same radiation protection requirements as for any other practices (that is, any practices involving exposure to artificial sources). As can be seen from Section 2.3, such requirements have not changed in any significant way from those set out in Safety Series No. 115.

It should be noted that, for practical purposes, everyday commodities such as foodstuffs, drinking water, fertilizer and construction material remain subject to the requirements for existing exposure situations, regardless of the activity concentration — for these materials, the application of the reference level for commodities (1 mSv or less per year) ensures that the level of protection of the public is similar to that for planned exposure situations. It is only the mechanism for controlling exposure that differs.

The decision process for determining whether an exposure is to be controlled as an existing exposure situation or a planned exposure situation is summarized in Fig. 1.

3.3. Exemption and clearance of material containing radionuclides of natural origin

When exposure to natural sources is, by exception, treated as a practice, the requirements for planned exposure situations are applicable, and these include requirements concerning exemption and clearance. For exemption, the two alternative approaches in Safety Series No. 115 remain in the new BSS draft, but some new criteria have been added:

(a) Exemption on a case by case basis: In terms of this approach, one of the following qualitative criteria for exemption has to be satisfied:
    (i) The radiation risks to individuals are sufficiently low as not to warrant regulatory control and the exempted practice or source is inherently safe (unchanged from Safety Series No. 115); or
    (ii) Regulation would provide no net benefit, in that no reasonable control measures would achieve a worthwhile return in reduction of individual doses or risks (a new criterion having its origin in the 1990 Recommendations of the ICRP, and which has particular relevance for exposure to natural sources);

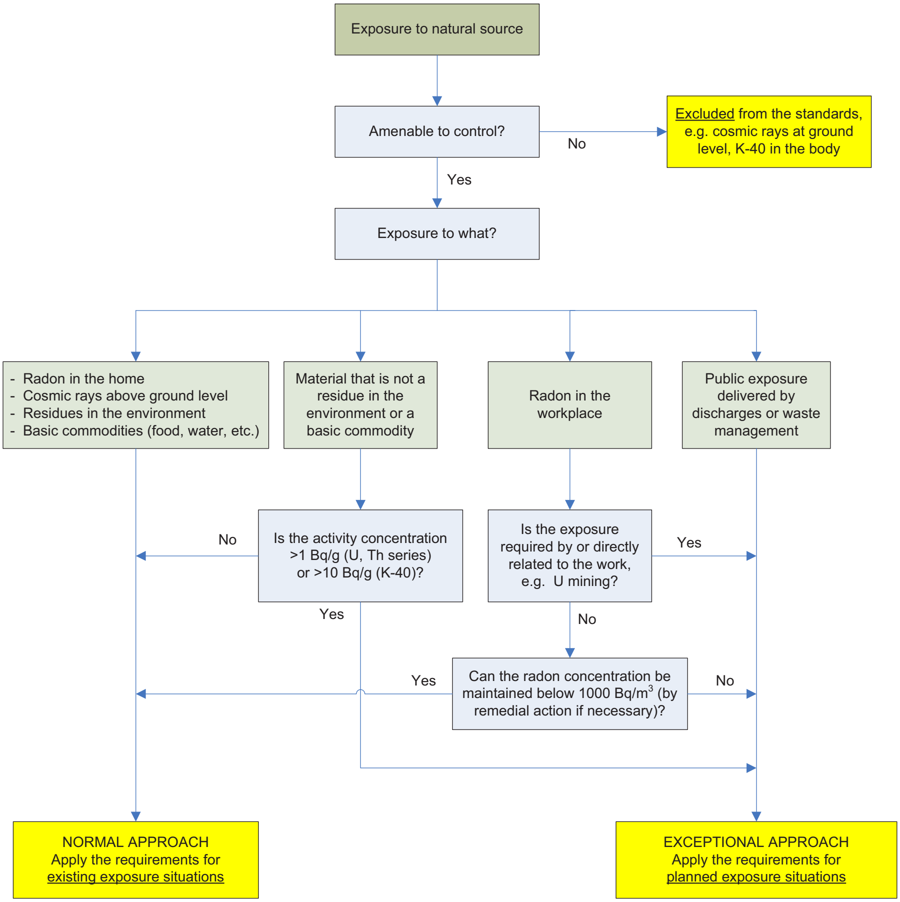

FIG. 1. Decision process for determining how to control exposure to natural sources.

(b) Automatic exemption without further consideration: In terms of this approach, the applicable criterion depends on which of two situations prevails:

(i) The first situation is one in which a radionuclide is incorporated into a consumer product or is used as a radioactive source (for example ^{226}Ra, ^{210}Po) or for its properties as a chemical element (for example thorium, uranium). In any event, the quantity of material is moderate (typically of the order of a tonne or less). The radionuclide is essentially treated in the same way as a radionuclide of artificial origin. The material is exempted without further consideration if, in all reasonably foreseeable situations, the effective dose expected to be incurred by a member of the public is of the order of 10 μSv or less in a year.[4] Conservative modelling shows that this dose criterion is met if the total activity of a given radionuclide present on the premises at any one time or the activity concentration used in the practice does not exceed the applicable exemption level given in Table 1 (representing no change from Safety Series No. 115).

(ii) Where a radionuclide is not incorporated for the purposes described in (a) above, implying that its presence is incidental, and where bulk quantities of material may be involved, the new BSS draft makes provision for exemption using a dose criterion commensurate with natural background levels, that is, about 1 mSv per year. This criterion is satisfied if the activity concentration does not exceed 1 Bq/g for uranium and thorium series radionuclides and 10 Bq/g for ^{40}K [7, 11]. However, these activity concentration values are generally too conservative to be of much practical use as exemption levels — material having such moderate activity concentrations would not even be subject to the requirements for planned exposure situations in the first place (see Section 3.2.1). Exemption at significantly higher

TABLE 1. EXEMPTION LEVELS FOR RADIONUCLIDES IN CONSUMER PRODUCTS OR USED AS A RADIOACTIVE SOURCE OR FOR THEIR PROPERTIES AS CHEMICAL ELEMENTS

Radionuclide[a]	Exempt activity (Bq)	Exempt activity concentration (Bq/g)
Th-230, Th-228	10 000	1
U-238, U-234, Ra-226, Pb-210, Po-210, Th-232	10 000	10
Bi-214, Ra-228, Ra-224, Pb-212, Bi-212	100 000	10
Ac-228	1 000 000	10
Pb-214, K-40	1 000 000	100
Bi-210	1 000 000	1000

[a] Progeny included in equilibrium: Ra-226, Pb-210, Th-228, Ra-224, Pb-212 and Bi-212 include all progeny; U-238 includes Th-234, Pa-234m; Th-232 includes Pa-234m; Ra-228 includes Ac-228.

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[4] In the new BSS draft, an additional dose criterion — an effective dose of 1 mSv or less in a year — has been introduced to take account of low probability scenarios, whereas the criterion based on collective dose (1 man·Sv) has fallen away.

> activity concentrations will generally be possible, even orders of magnitude higher in some situations. Since the relevant activity concentration value will depend strongly on the exposure scenario, an assessment against the 1 mSv criterion for the particular exposure scenario is required.

The decision process to determine whether a practice or a source within a practice should be exempted is illustrated in Fig. 2.

The two alternative approaches for deciding upon exemption, as described in (a) and (b) above, apply also to clearance. For the case by case approach, the criteria are essentially the same as those for exemption. For automatic clearance without

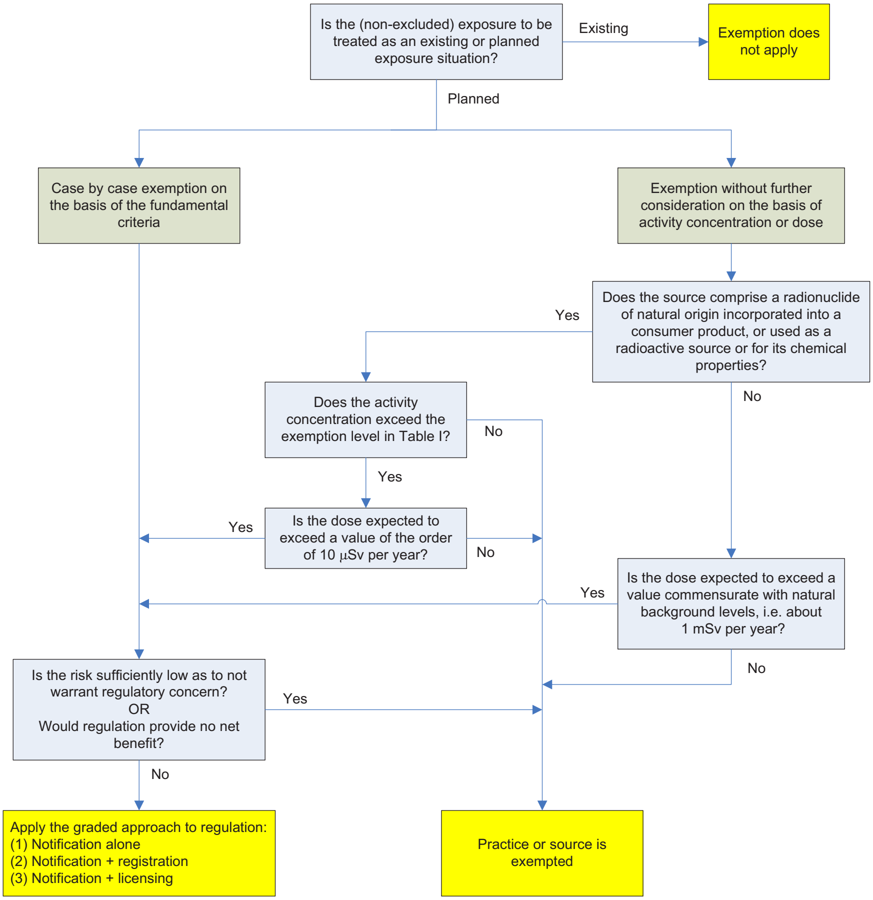

FIG. 2. Decision path for exemption with respect to exposure to natural sources.

TABLE 2. NUMERICAL CRITERIA FOR EXEMPTION AND CLEARANCE

	Criterion		
	Maximum effective dose (mSv per year)	Maximum activity (Bq)	Maximum activity concentration (Bq/g)
Exemption:			
— Radionuclides incorporated into consumer products, or used as a radioactive source or for their properties as chemical elements[a]	~0.01	10^4–10^6 depending on radionuclide	1–1000 depending on radionuclide
— Other situations	~1	—	—
Clearance	—	—	1 (U, Th series) 10 (K-40)

[a] Only one of the three criteria has to be satisfied.

further consideration, the clearance criteria are that the radionuclide activity concentration must not exceed 1 Bq/g for uranium and thorium series radionuclides and 10 Bq/g for ^{40}K, irrespective of the quantity of material. These clearance criteria were established in 2004, but were published only at the Safety Guide level [7].

A summary of exemption and clearance levels is given in Table 2.

4. CONCLUSIONS

The new BSS draft follows closely the 2007 Recommendations of the ICRP. The structure has been simplified and a set of overarching requirements has been incorporated throughout the document. This should improve the user friendliness. In revising the requirements for exposure to natural sources, greater use has been made of quantitative criteria rather than qualitative criteria. While there are still some complexities in dealing with exposure to natural sources, the requirements in this area should be easier to follow and should result in a more harmonized approach to regulatory control, particularly with respect to exemption and clearance. For existing exposure situations, the new ‘reference level’ approach of the ICRP has been adopted, and this will have the intended effect of increasing the level of protection against exposure to natural sources by removing any lower bound on the optimization process. In the case of reference levels for indoor radon, the new values of radon concentration reflect the latest ICRP recommendations in terms of which the dose per unit activity concentration of inhaled radon is considered to be higher than previously assumed.

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RADIOACTIVITY ASSESSMENT IN PHOSPHATE INDUSTRY FACILITIES: THE OCP GROUP’S EXPERIENCE

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Abstract

Phosphorus (usually expressed as P_2O_5) is one of the main elements used in fertilizer manufacture. Twenty five percent of the fertilizer consumed in the world uses P_2O_5. Phosphate ore is a vital raw material for the production of fertilizers. It contains, in addition to P_2O_5, a multitude of other elements, some of which are radioactive. The radionuclide decay chains are approximately in equilibrium. During fertilizer production, the chemical transformation causes a new distribution of radionuclides among the different phases: phosphoric acid, phosphogypsum, fertilizers and deposits (scales); this depends on the chemical behaviour of each element in phosphoric acid. Studies have shown that the radioactivity content of deposits inside tubular and other process equipment (including pipes carrying phosphoric acid) is mainly due to the presence of radium and its decay products (^{226}Ra, ^{214}Pb and ^{214}Bi). Hence, radiation doses increase in the vicinity of this equipment. In order to control exposure, regular measurement campaigns have been undertaken across the entire facility. This has allowed a detailed map of dose distributions to be established. A washing solution has been developed that effectively dissolves radioactive scales, thus substantially reducing radiation doses down to low levels. This paper describes the approach followed and the results achieved.

1. RADIOACTIVITY IN PHOSPHATES

1.1. Natural substitutions in the crystal structure

The impurities in phosphate rock are extremely varied in type and concentration. They are found in either the apatite structure, whose crystalline sites allow a substitution, or as crystalline or amorphous compounds in phases formed in the gangue of the phosphate ore. Minerals of the apatite group have the general composition $M_{10}(ZO_4)_6X_2$, with the most common cations being M = Ca and Sr; Z = P, As and Si; and X = F, OH, Cl and CO_3^{2-}. Such minerals are exceptionally tolerant to structural distortions, making the substitution of a very large number of elements in its structure possible (see Fig.1).

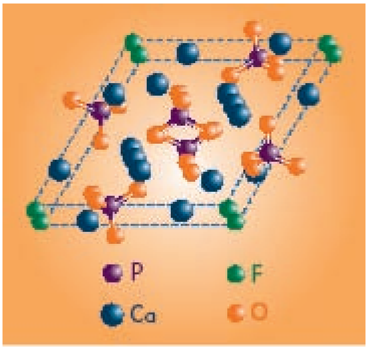

FIG.1: Projection of apatite $Ca_{10}(PO_4)_6F_2$ *hexagonal mesh.*

In natural apatites, the largest variety of substitutions is observed in sites with cations having the same valency as calcium, but whose ionic radii may be different [1]. These include Sr^{2+}, Mn^{2+}, Pb^{2+}, Ba^{2+}, Eu^{2+}, Cd^{2+}, Fe^{2+}, Mg^{2+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+} and Sn^{2+}. Usually the concentrations of these elements are around tens to hundreds of parts per million. However, cations with other valencies can also be substituted. These include monovalent Na^+, K^+, Li^+ and Rb^+; trivalent Y^{3+} and Al^{3+}; and tetravalent U^{4+} and Th^{4+}.

1.2. Sources of radioactivity in phosphates

Most phosphates contain between 40 and 150 ppm uranium in tetravalent (+IV) and hexavalent (+VI) forms. The latter form occurs more commonly in phosphates that have been oxidized [1]. The behaviour of uranium in phosphate ore depends on the oxidation or reduction state. In an oxidizing environment, the uranium is in the tetravalent form +VI (the uranyl ion UO_2^{2+}). It is the most stable and mobile form. Hexavalent uranium is far more soluble than tetravalent uranium. It forms complexes, the most common being uranylcarbonates and uranylsulphates.

Radionuclides in the uranium and thorium decay series are the main source of radioactivity in phosphate ore [2]. Natural uranium comprises three isotopes (^{234}U, ^{235}U and ^{238}U), which decay by emitting alpha and beta radiation to produce various decay progeny. Phosphate ore also contains radionuclides in the ^{232}Th decay series and ^{40}K. The ^{238}U decay series is the most important source of radioactivity. It represents more than 95% of the radionuclide content of Moroccan phosphate ore. The radionuclides in the ^{238}U and ^{232}Th decay series, together with the type of radiation emitted, are shown in Figs 2 and 3.

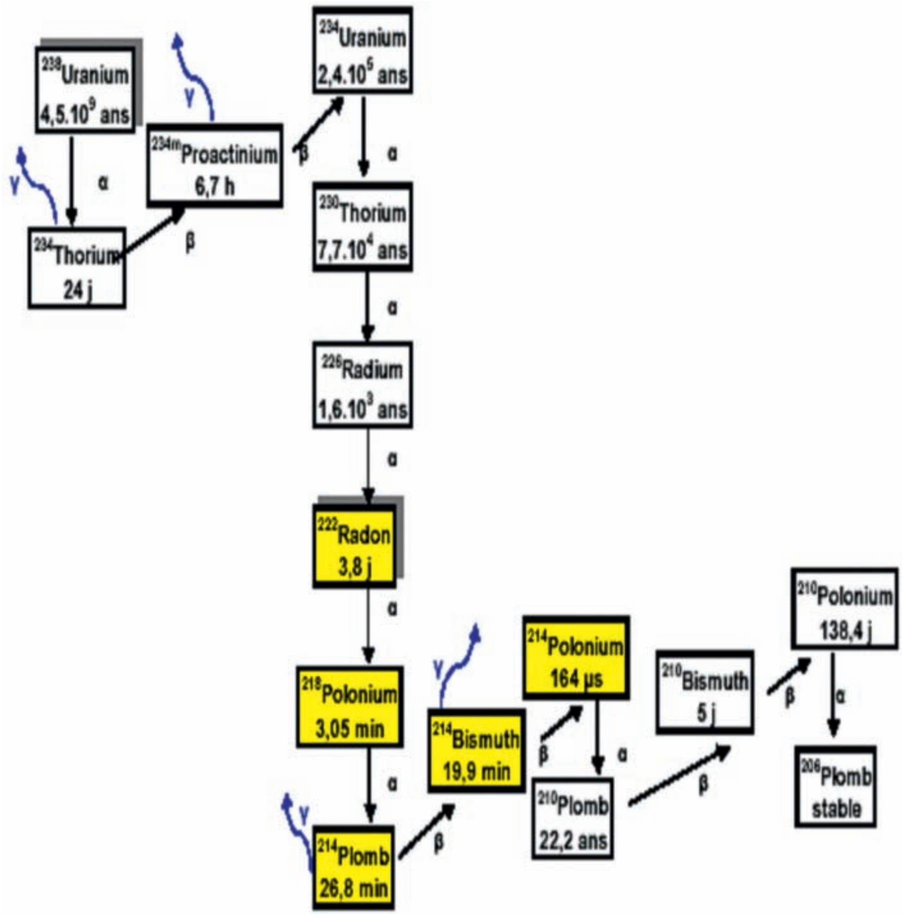

FIG. 2. Uranium-238 decay series.

Because of the age of phosphate ores, these decay series are generally in equilibrium. However, factors such as the release of radon and the elutriation of uranium by rainwater or groundwater can lead to small degrees of disequilibrium.

2. BEHAVIOUR OF RADIONUCLIDES DURING PHOSPHATE PROCESSING [2]

In most types of phosphate rock, uranium is approximately in equilibrium with its decay progeny. This equilibrium is disturbed during the production of phosphoric acid and fertilizer and the radionuclides become distributed in different ways among the various process materials (acids, gypsum and deposits).

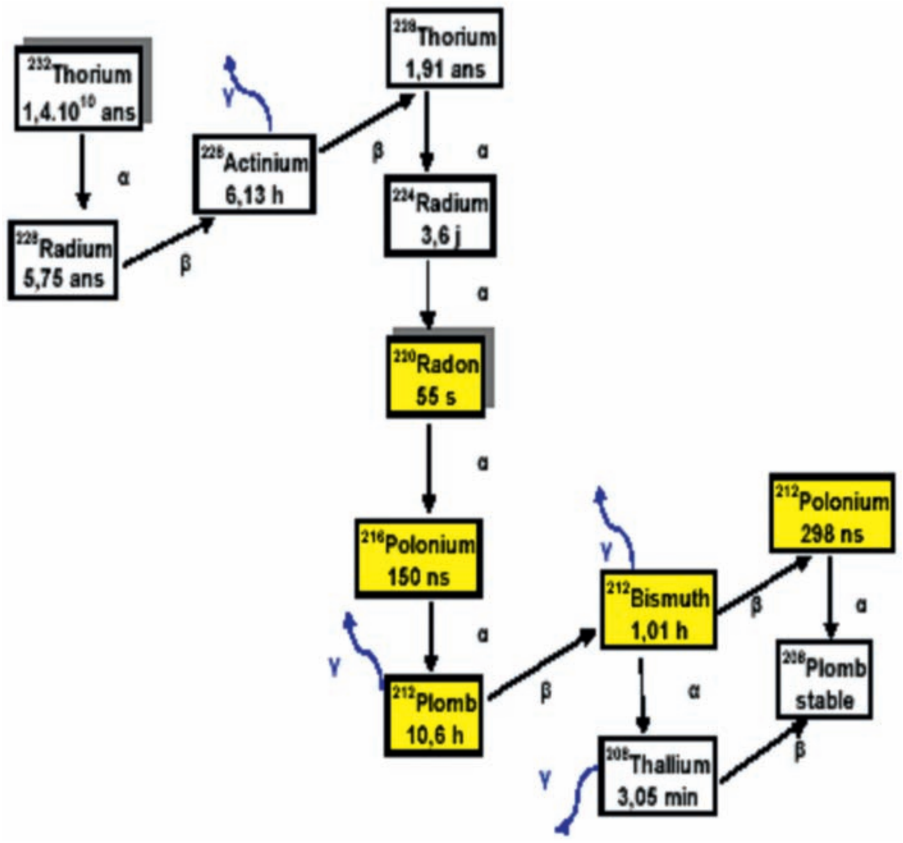

FIG. 3. Thorium-232 decay series.

Studies have been undertaken to determine the behaviour of radionuclides during the reaction, filtration and concentration steps of phosphoric acid production, as well as during the production of fertilizer from phosphoric acid. These studies were concerned with the distribution of U^{-238} series radionuclides (^{238}U, ^{234}Th, ^{226}Ra, ^{214}Pb and ^{214}Bi), which are the source of radioactivity in Moroccan phosphate. The studies led to the following conclusions:

(a) The phosphate rock used for the production of phosphoric acid has a radionuclide activity concentration of about 1 Bq/g.
(b) In all types of phosphate rock, the radionuclides are in equilibrium.
(c) The washing of phosphate rock to remove the gangue results in an increase in the radionuclide concentration, indicating that the radionuclides are bound to the apatite and not to the gangue.

(d) The radioactivity content of phosphoric acid is mainly due to the presence of ^{238}U, ^{230}Th and ^{234}Th. This is a result of the high solubility of these radionuclides in phosphoric acid. On the other hand, the acid contains essentially no ^{226}Ra, ^{214}Pb or ^{214}Bi.

(e) The radioactivity content of the phosphogypsum residue is mainly due to the presence of ^{226}Ra. Most of the ^{226}Ra, ^{214}Pb and ^{214}Bi activity ends up in the phosphogypsum, scales and acid solids. Radium-226 is deposited in the phosphogypsum by the mechanism of co-precipitation with barium sulphate. The formation of $RaSO_4$ in phosphogypsum is thus only possible in the presence of barium.

(f) The distribution of ^{238}U between the phosphoric acid and the phosphogypsum is the same, regardless of whether the dihydrate (DH) process or the hemidihydrate (HDH) process is used for the production of phosphoric acid.

(g) When phosphoric acid is concentrated, the concentrations of uranium and thorium series radionuclides are also increased. However, small amounts of these radionuclides migrate to the strong acid solids.

(h) The radioactivity in MAP, NPK and DAP fertilizers is due to the presence of uranium and thorium isotopes. This is explained by the high solubility of these radionuclides in the phosphoric acid used for the production of these fertilizers.

(i) TSP fertilizer is characterized by the presence of five radionuclides from the ^{238}U decay series.

(j) The radioactivity content of scales and other solid deposits is mainly due to the presence of radium and its progeny.

(k) The radioactivity content of filter separator scales is due to the accumulation of ^{226}Ra, ^{214}Pb and ^{214}Bi.

3. DOSES RECEIVED BY WORKERS

3.1. Assessment by calculation

In order to classify the phosphate industry according to the NRPB[1] classification scheme [3], the annual effective doses received by workers in the plants were calculated. The conditions taken into account were:

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[1] NRPB refers to the former National Radiological Protection Board in the United Kingdom (now the Radiation Protection Division of the Health Protection Agency).

(a) The generic conditions published in NRPB reports;
(b) The actual environmental conditions in the facilities considering the five scenarios developed by the NRPB.

The calculations were based on the models and equations of the International Commission on Radiological Protection [4] and guidance in Ref. [5] related to the 1996 Euratom Directive [6]. The work areas considered were:

(i) The phosphate handling plant;
(ii) The fertilizer plant;
(iii) Phosphogypsum filtration;
(iv) Phosphoric acid.

The results are shown in Tables 1–4. For the phosphate handling and fertilizer plants, under the NRPB conditions the classification system would have required a high level of radiological control in some areas (Zone 3 and workers classified as category A). Under the actual industrial work conditions, different parameters were varied to take into account different cases. The annual doses were in the range 0.34–2.35 mSv at plant 1 and 1.24–2.71 mSv at plant 2. In these cases the maximum annual dose remained in the region of 1 mSv. For the phosphogypsum filtration and phosphoric acid scenarios, the annual dose was less than 1 mSv under both NRPB and actual conditions. The NRPB classification system would not have imposed any radiological control (Zone 1 and workers classified in the B category). Overall, when taking into account real working conditions, the calculations suggest that the phosphate industry is classified at the lower end of Zone 2.

TABLE 1. CALCULATED ANNUAL EFFECTIVE DOSE, PHOSPHATE HANDLING PLANT

		Annual effective dose (mSv)	Annual equivalent dose to the skin (mSv)	NRPB classification
NRPB conditions	Plant 1	9.05	1.58	Zone 3
	Plant 2	9.28	1.53	Zone 3
Actual working conditions at OCP	Plant 1	0.5–1.34	0.08	Zone 1–2
	Plant 2	1.24–2.39	0.46	Zone 1–2

TABLE 2. CALCULATED ANNUAL EFFECTIVE DOSE, FERTILIZER PLANT

		Annual effective dose (mSv)	Annual equivalent dose to the skin (mSv)	NRPB classification
NRPB conditions	Plant 1	1.39	0.33	Zone 2
	Plant 2	1.48	0.31	Zone 2
Actual working conditions at OCP	Plant 1	0.34–2.35	0.08	Zone 1–2
	Plant 2	1.24–2.71	0.46	Zone 1–2

TABLE 3. CALCULATED ANNUAL EFFECTIVE DOSE, PHOSPHOGYPSUM FILTRATION

		Annual effective dose (mSv)	Annual equivalent dose to the skin (mSv)	NRPB classification
NRPB conditions	Plant 1	0.05	0.004	Zone 1
	Plant 2	0.05	0.004	Zone 1
Actual working conditions at OCP	Plant 1	0.12–0.45	0.01	Zone 1
	Plant 2	0.7	0.004	Zone 1

TABLE 4. CALCULATED ANNUAL EFFECTIVE DOSE, PHOSPHORIC ACID

		Annual effective dose (mSv)	NRPB classification
NRPB conditions	Plant 1	0.01	Zone 1
	Plant 2	0.003	Zone 1
Actual working conditions at OCP	Plant 1	0.01	Zone 1
	Plant 2	0.009	Zone 1

3.2. Assessment by site measurements [2]

In order to verify the NRPB classification and to determine the annual doses received by workers under real working conditions, radioactivity measurements were made at OCP’s industrial sites (mining facilities and fertilizer plants). The measurements made at the fertilizer plants covered the following workplaces: phosphate storage areas, fertilizer storage areas, granulation plants, phosphoric acid storage areas and phosphoric acid filtration areas. The measurements were made using instruments that could detect radon and gamma exposure simultaneously. The measurement programmes lasted more than one year and took account of external (gamma) radiation, the inhalation of radon gas and the ingestion and inhalation of dust. The annual effective doses were calculated using the actual times of exposure in the workplaces concerned. The workplaces were classified on the basis of the normal doses likely to be received by the workers. The results are shown in Tables 5 and 6.

TABLE 5. WORKPLACE CLASSIFICATION ACCORDING TO WORKER DOSES ASSESSED FROM SITE MEASUREMENTS, MINING FACILITIES

	NRPB classification
Storage	Zone 1–2 (1.40 mSv/a)
Screens	Zone 1
Furnace	Zone 1
Train loading	Zone 1–2 (2.3 mSv/a)
Dragline	Zone 1
Excavation	Zone 1

4. DISSOLUTION OF RADIOACTIVE SCALES

During phosphate processing, scales are formed in parts of the plant. These scales contain radionuclides from the uranium decay series and it is for this reason that some work areas were classified as Zones 1 and 2. The chemical removal of these scales provides an opportunity for reducing the doses received by workers.

4.1. Laboratory tests

Laboratory tests were undertaken to evaluate how best to reduce these scales [2], with the intent of determining an efficient solubilization process for the radionuclides incorporated in the scales. The experimental method is illustrated in Fig. 4. The sample (a piece of pipe with scale deposit) was immersed in the washing solution and maintained at a temperature of 60°C for 2 h. An agitator created turbulence within the pipe sample. The dose rates were measured before

TABLE 6. WORKPLACE CLASSIFICATION ACCORDING TO WORKER DOSES ASSESSED FROM SITE MEASUREMENTS, FERTILIZER PLANTS

	NRPB classification
Phosphate warehouse	Zone 1–2 (1.8 mSv/a)
NEF	Zone 1–2 (1.5 mSv/a)
Phosphate hopper	Zone 1–2 (1.8 mSv/a)
Control room	Zone 1
Fertilizer storage (cabin)	Zone 1
Fertilizer storage (walkway)	Zone 1
Fertilizer conveyor	Zone 1
Unit 18 (top of scraper)	Zone 1
Warehouse (bottom of scraper)	Zone 1–2 (1.3 mSv/a)
Granulation plant (grinding)	Zone 1
Filter (strong acid)	Zone 2 (2 mSv/a)
Filter (control room side)	Zone 1
Filter (above separator)	Zone 1
Storage, 29% phosphoric acid	Zone 1
Storage, 54% phosphoric acid	Zone 1
Thickener, 29% acid	Zone 1
Thickener, 54% acid	Zone 1
Separator	Zone 2 (2.6 mSv/a)

and after each test on the sample with a portable detector. Several solutions were tested and the best results were achieved using a solution known as BEL10.

4.2. Industrial application tests, reaction and filtration area

To optimize the consumption of the reagents and to enable recirculation of the washing solution, some flanges and additional pipes were installed in the wash water recovery network. The washing circuit is shown schematically in Fig. 5 and the results are given in Table 7. The industrial tests resulted in an 8–35 % reduction in dose rates in the area of study. The implementation of the proposed solution would allow the phosphoric acid circuit to be reclassified as Zone 1.

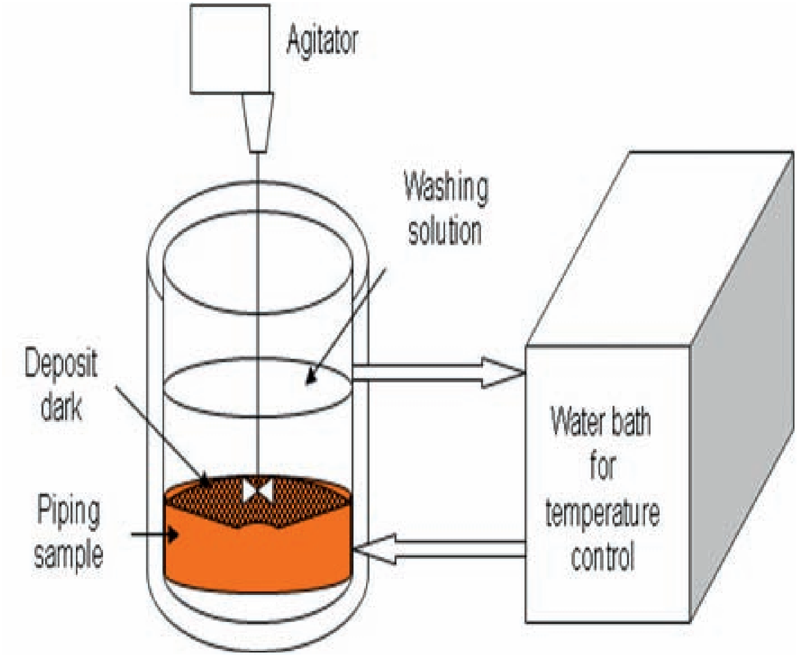

FIG.4. Equipment used for the laboratory scale experiment.

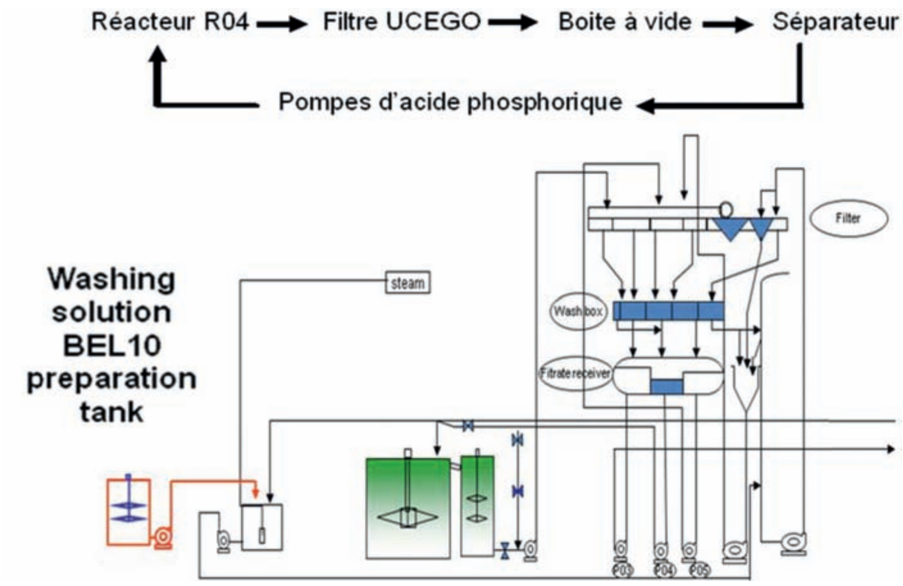

FIG. 5. Washing circuit.

TABLE 7. EFFECT OF THE WASHING SOLUTION ON THE REDUCTION OF DOSE RATES

		Reduction in dose rate (%)	
		Line 1	Line 2
Filter	Gypsum hopper	28	26
	Filter table	35	19
Filtrate receiver	Filtrate sectors	8–25	12–20
	Manhole	12–15	16–18
	Phosphoric acid piping	18–23	24–31
Collector	Piping	12	17
	Cloth wash piping	25	22
Pipes	Phosphoric acid piping	14–17	19–32
	Phosphoric acid piping	11–32	11–31

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OPERATIONAL RADIATION PROTECTION

(Topical Session 1)

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Invited Paper

THE USE AND MANAGEMENT OF NORM RESIDUES IN PROCESSING BAYAN OBO ORES IN CHINA[1]

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Abstract

Bayan Obo mine has a deposit of iron and rare earth ores that is renowned as a large rare earths deposit. The ores are rich in radioactive elements, with a 0.01–0.05% concentration of ThO_2 and a 0.0005–0.002% of U_3O_8. The deposit has been mined for more than 50 years. The ores are transported by train to the refinery plants in Baotou to process for products of iron and steel, rare earths and their compounds. Meanwhile, a large amount of NORM residue produced is being regulated and controlled. At present, about 560 million t of waste rock produced are stored in the on-site waste rock dumps around the open pits, 149 million t of tailings are stored in a tailings pond, about 55 million t of ferrous slag are stored in a ferrous slag dump and 437 300 t of rare earth slag are stored in a radioactive waste storage facility. Most of the wastewater, after being treated, is discharged into the tailings pond and then pumped to the milling plant for reuse. Waste gas, after dust removal, is discharged to the environment. Any utilization of NORM residues must comply with radiological regulations in China. A substantial amount of blast furnace iron slag has been made into cement, concrete and bricks or used directly for highway construction. The development and exploitation of NORM residues is of importance for waste minimization. However, it raises serious environmental concerns. How to implement the ALARA principles of optimization, to safely utilize the residues and to reduce the waste are therefore important topics in the management of NORM residues.

1. INTRODUCTION

The worldwide industry reserve of rare earths (REs) is about 112.7 million t (expressed as rare earth oxides), whereas China possesses a reserve of 43 million t, the largest in the world. The Bayan Obo deposit has 35 million t, accounting for 81.4% of the total amount in China. The remaining 8 million t are distributed in Sichuan, Shandong, Jiangxi, Guangdong, Taiwan and other provinces [1]. Bayan Obo mine, comprising the Main Mine, East Mine and West Mine, contains minerals such as iron, rare earth elements, thorium and niobium. The Main Mine and East Mine have reserves of 600 million t of ore containing about 34% iron, 5% REO and 0.032% ThO_2. The deposit at the West Mine has 800 million t of ore containing 33.15% iron and a low content of REs, thorium, phosphorus and fluorine [1]. About 276 million t of ore had been mined by the end of 2006. About 10 million t/a of ore are currently mined and supplied as raw materials for steel and rare earth element production to BTISP (Inner Mongolia BaoTou Iron and Steel Plant (Group Ltd.)) and as concentrated raw materials to other rare earth plants in Baotou.

BTISP, which was founded in 1954, is located in western Baotou 150 km south of Bayan Obo. Ore from Bayan Obo is transported by train to BTISP for processing. BTISP produced only iron and steel products at its early stage. It has been producing RE products from RE concentrates in conjunction with iron and steel since 1974. The BTISP processes 12 million t/a of ore from the Bayan Obo mine, produces 9 million t/a of iron and steel and more than 7000 t/a of rare earths (expressed as rare earth oxide (REO) equivalent) [2].

The ores contain elevated concentrations of thorium, so mining and processing have a certain radiological impact on workplaces and the environment. An investigation and assessment of radioactive pollution from the exploitation of the Bayan Obo deposit was conducted and funded by the Ministry of Environmental Protection of China and the China Aero Geophysical Survey and Remote Sensing Center for Land and Resources. This paper presents some results of this project.

2. MINING AND PROCESSING

The Main Mine and East Mine are designed for a production capacity of 525 million t of ore, made up of 337 million and 188 million t of ferrous ore from the Main Mine and East Mine, respectively. The mining areas are about 1520 × 1080 m and 1400 × 1020 m for the Main Mine and East Mine, respectively. Annual mining is 10 million t of ore, declining year by year. The engineering design and mining of the West Mine started in May 2006. It has been developed into a large open pit mine 4600 m long and 1000–1200 m wide. The present production of ore is expected to be 3 million t/a, and is increasing as time goes on.

A large amount of waste rock is generated during mining, and is deposited on dumps on the mine site. About 10 million t of waste rock are produced annually. Ore from the open pit is conveyed initially to a crushing facility at Bayan Obo. The crushed ore is then transported by train to BTISP for sorting and smelting to iron and RE concentrates. The annual production of iron concentrate and RE concentrate is about 4.5 million t and 100 000 t respectively [3]. The iron concentrate contained 0.024% thorium in the early stages; however, it has been reduced to 0.0073% after technical improvements. The rare earth concentrate contains 0.2% thorium. The generation of tailings is about 6.55 million t/a with a thorium content of 0.048%, which is discharged into the Baogang tailings pond for storage [4, 5].

All the iron concentrate from Bayan Obo ore is reserved for BTISP to produce steel products, accounting for 40% of total production. Iron and steel products are produced from iron concentrate through processes of sintering, blast furnace melting and conversion. The process generates a large amount of blast furnace slag. BTISP currently produces 9 million t of iron and steel, yielding 3.55 million t of ferrous slag annually [3], along with dust particulates and liquid effluents. The ferrous slag is conveyed to a slag dump for storage. The liquid effluents are discharged into the tailings pond.

About two thirds of the rare earth concentrates from Bayan Obo are reserved for the RE plants in Baotou, the rest for plants outside Baotou. There are altogether 15 RE plants in Baotou, 13 of them located in the urban area. The diversified products include RE oxides, RE chlorides, RE carbonates and alloy products. Chemical processes include both acidic and alkaline methods, but the acidic method is used most widely. An amount of 60 000 t/a of RE slag is produced and disposed of in the Baotou radioactive waste storage facility [4]. Exhaust gas, after being treated by gas cleaning, is discharged to the environment. Further details concerning the extraction and processing of iron and steel and RE products from the ore can be found in Refs [6, 7].

3. MANAGEMENT AND UTILIZATION OF WASTES

Radiological safety in the exploitation of Bayan Obo ore and the utilization of NORM residues is ensured through compliance with the national regulations, as well as relevant IAEA standards [8–11]. The iron and steel products contain only trace levels of radioactive elements or none at all. During the processing or smelting of iron and steel, the thorium content of the ore is redistributed into solid waste in the form of slag (96–98%), exhaust gas (0.1–0.5%) and liquid effluent (0.6–2.0%) [3]. However, the situation is quite different for RE products, for which the thorium content is 0.04–0.24% for RE alloys, about 0.045% for RE chlorides and 0.0069% for RE oxides. A flow chart for the generation of wastes in the mining and processing of Bayan Obo ores is shown in Fig.1. The total production of solid wastes at the end of 2006 is shown in Table 1.

TABLE 1. INVENTORY OF SOLID NORM RESIDUE [3]

Slag type	Production (t)		Storage location
	2006	Total, to 2006	
Tailings	6 546 700	149 337 000	Baogang tailings pond, Bayan tailings pond
Alloy slag	5 800	963 000	Baogang alloy slag dump
Blast furnace slag	2 900 000	55 025 000	Baogang ferrous slag dump
RE	60 900	261 000	Baotou radioactive waste storage facility

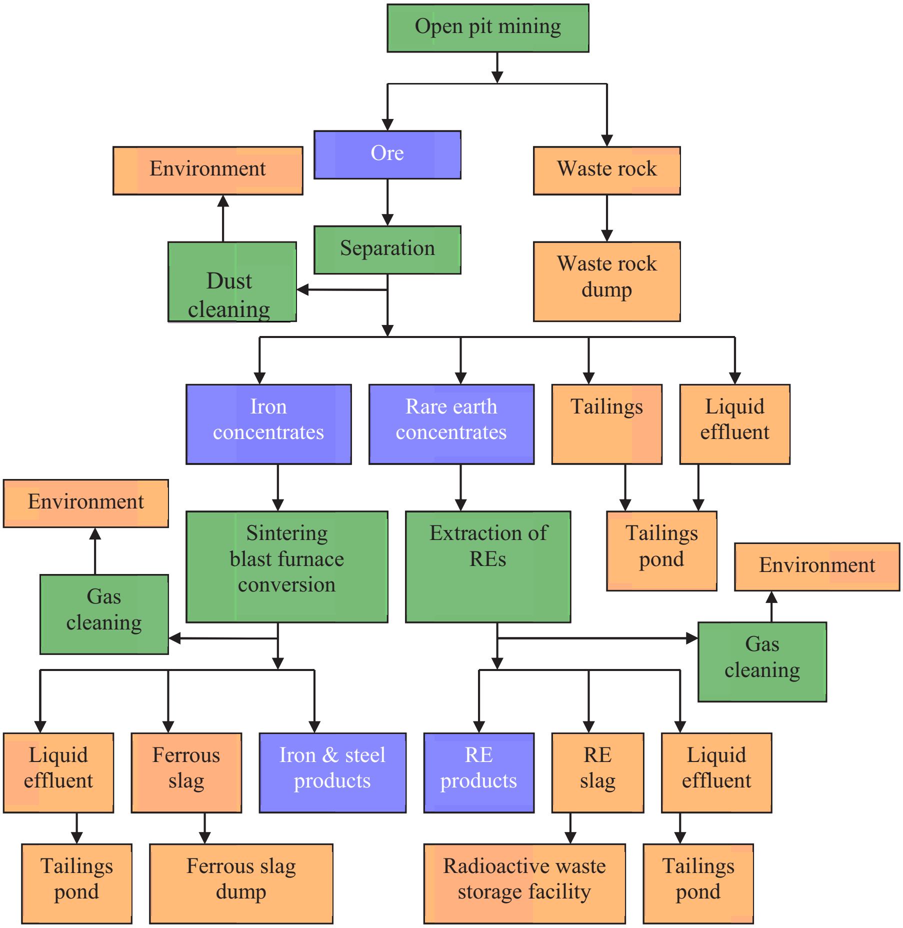

FIG. 1. Major flow chart and pollutant production of Bayan Obo ore exploitation.

3.1. Solid waste

3.1.1. Waste rock

The waste rock comprises topsoil, barren rock and low grade mineralized rock from around the orebody. This waste is mostly in the form of large coarse blocks that are piled up in a dump near the mining area according to the design requirements. The low grade mineralized rock may be recycled once the sorting technique is improved in the future. The topsoil and host rock may be returned to the excavation as backfill when the mining is finished. The Bayan Obo deposit has been exploited for more than 50 years, so a total amount of 560 million t of waste rock is piled up in the waste rock dumps. Sprinklers are deployed to spray water regularly to mitigate dust at the mining site for improvement of the environment and reduction of inhalation of dust containing radioactive elements [4].

3.1.2. Slag

The slag is mainly composed of the tailings from the sorting, the ferrous slag from the ferrous smelting and the RE slag from the RE extraction. Among these, the tailings component is the largest, amounting to 149 million t at the end of 2006 and occupying an area of 11 km^2 The ferrous slag is the second largest, amounting to 55 million t and covering an area of 1.27 km^2. The RE slag, although in a smaller quantity (437 300 t), is strictly regulated because its radioactivity approaches the level of low level radioactive solid waste [4].

The tailings have an elevated ^{232}Th content of about 1.6 Bq/g and constitute a major pollution source. In order to reduce the airborne dispersion of tailings, some measures have been introduced. For example, water spraying is applied to wet the surface of the tailings regularly, the tailings pond is covered with waste water from ore processing, and trees are planted in the area surrounding the tailings pond.

The ^{232}Th activity concentration of ferrous slag is in the range of about 0.5–1.6 Bq/g [5], which is partly above the specified exemption level of 1 Bq/g [7, 9]. The slag containing elevated levels of thorium and usable iron is stored in the ferrous slag dump, which is isolated from the surrounding environment.

The gross alpha activity concentration of the alloy slag is in the range of 2.9–5 Bq/g [2]. The amount of alloy slag in the stockpile is 963 000 t, which is mostly stored in the alloy slag dump beside the ferrous slag dump for overall management.

The acidic method is applied to extract RE. The gross alpha activity concentration of acidic process slag is 80–200 Bq/g, which exceeds the activity level of low level radioactive solid waste. Hence, it is required to store it in the Baotou radioactive waste storage facility. The gross alpha activity concentration of alkaline process slag is 1000 Bq/g, which is higher than that of acidic process slag. All alkaline slag is recovered and used in the acidic process.

3.2. Liquid effluents

Many of the liquid effluents are produced by the wet process method and are discharged into the tailings pond after treatment. The total liquid effluent is about 17 million m^3 with a gross alpha activity concentration of 5.39 Bq/L and a gross beta activity concentration of1.58 Bq/L [12]. The liquid effluent discharged into the tailings pond was recorded at 65 875 000 t in 2006, of which 64 572 100 t have been recovered. The effluent contains, in addition to thorium radionuclides, fluorine, nitrogen (as ammonia) and a large amount of sulphate and chlorine ions.

3.3. Gaseous effluents

The gaseous effluents include:

(a) Airborne dust produced by primary crushing and sorting;
(b) Exhaust gases from a variety of processes including roasting of the concentrate, sintering, the blast furnace, RE acidic method roasting and RE alloy smelting.

It is reported in Ref. [12] that the activity concentrations in the airborne dust from crushing and sorting were 1.3–1.9 Bq/g for ^{232}Th and 1.2–2.0 Bq/g for ^{228}Ra. The activity concentrations in the dust from sintering were 0.05–0.16 Bq/g for ^{232}Th and 0.12–0.20 Bq/g for ^{228}Ra. The dust from steel smelting had activity concentrations of 0.02–0.12 Bq/g for ^{232}Th and 0.02–0.11 Bq/g for ^{228}Ra. The Baotou RE roasting operation processed 102 400 t of concentrate in 2006, during the course of which 2460 million m^3 of exhaust gas were generated, containing 78.6 MBq of ^{232}Th [4].

3.4. Utilization of wastes

3.4.1. Recovery of acid

Extraction of RE by the acidic method produces a large amount of effluent which has a certain impact on the tailings pond. Some technical improvements were made to recover the sulphuric acid from the scrubbed effluent in 2004. With an annual recovery of 45 000 t of sulphuric acid, the condition of the water has been improved since then.

3.4.2. Recovery of iron

The earlier technique of steel smelting used at BTISP was not as advanced as at present. The ferrous slag contained some residual iron. The present recovery process uses a magnetic sorting method to recover the residual iron; the iron-free residue is used for construction or as raw material.

3.4.3. Uses of waste rock

Waste rock can be used in different construction materials. However, since the radioactivity levels of the waste rock vary significantly in mining locations, classification according to these levels is needed in advance. The radioactivity level is lower for the upper layer of rock and the host rock, while rock near the ore boundary has a higher radioactivity content. These wastes are now mainly used for road construction and for the embankment of the new Bayan Obo tailings pond, but they contribute only a small amount to the total used in these applications.

3.4.4. Uses of slag

The extraction of thorium has been achieved successfully in the laboratory but has not been applied on an industrial scale. RE residues are stored as a reserve in the radioactive waste storage facility. Ferrous slag is used to produce qualified construction products or as raw materials, aside from the recovery of waste iron mentioned above. The blast furnace slag is used in the following ways:

(a) Some is sold to cement factories, brickyards and other construction material manufacturers after water quenching (and is therefore known as water quenching slag);
(b) Some is delivered to a ferrous slag dump for storage; it is crushed and then sorted by magnetic separators or classified for recovery of waste iron that will be used in steel smelting;
(c) The remainder is used for filling, construction or brick manufacturing.

Out of a total of 3 552 530 t of blast furnace slag produced at BTISP in 2007, 750 480 t were consumed as water quenching slag and 701 776 t were consumed as ferrous slag [3]. Some major enterprises are listed in Table 2. The consumption of the slag accounts for only half of the production based on the recent utilization rate. Therefore, BTISP is giving consideration to increasing the production and sales of economical, safe and environmentally qualified products.

The usual way to produce construction materials from blast furnace slag is to mix the slag with low activity material such as fly ash in a ratio such that the activity concentration is sufficiently reduced to allow conversion into construction materials meeting radiological requirements. A formula from a manufacturer for 52 208 m^2 of paving bricks and 32 144 m^3 of house bricks is: 60–70% steel slag, 10–15% blast furnace water quenching slag, about 10% fly ash and 12–17% cement. The products meet the radiological requirements for construction materials.

TABLE 2. MANUFACTURING ENTERPRISES USING FERROUS SLAGS IN 2007

	Production	Activity concentration (Bq/g)		
		Ra-226	Th-232	K-40
Brick factory	52 208 m^2 paving bricks 32 144 m^3 house bricks	0.051	0.212	0.123
Cement factory	600 000 t of cement	0.0836	0.3309	0.4292
Cement factory	378 000 t of 425# and 325# cement	0.0246	0.2401	0.3719

4. EXPOSURES AND ENVIRONMENTAL IMPACT

4.1. Environmental radiation in the Bayan Obo mine area and its surroundings

4.1.1. Radiation levels

Radiation levels were determined by airborne and in-situ gamma spectrometry and dosimetry [3]. The background absorbed dose rate was 0.085 μGy/h. Higher levels were found mostly in the mining area and its surroundings, including the mining sites, some of the office buildings and the disposal sites; these locations are considered as ‘high background radiation areas’. Typical levels were in the range 0.2–0.8 μGy/h over an area of about 55 km^2, with levels of 0.6–2 μGy/h at the mining sites and 0.4–0.8 μGy/h at the disposal sites. The levels in some areas with low grade mineralized rocks were up to 1.2 μGy/h. Elevated levels in areas including the new RE plant and tailings pond, sintering plant and iron plant were typically in the range 0.3–0.5 μGy/h.

Because of the influence by open pit mining, the absorbed dose rates in areas in the leeward direction from the mine, including the Bayan Obo City area, were in the range 0.1–0.15 μGy/h, averaging 0.121 μGy/h, which is 50% higher than the normal background value. The contaminated soil is distributed mostly in the upper 10 cm layer; the activity concentration of thorium in this layer was about 0.08–0.12 Bq/g.

Elevated radiation levels were also observed with respect to thoron in air [13]. The potential alpha energy concentrations (PAECs) of ^{220}Rn progeny were 199.4 MeV/L (0.032 μJ/m^3) in the urban areas and 226.1 MeV/L (0.036 μJ/m^3) at the disposal site.

4.1.2. Exposures

(a) Occupational exposure: It is reported in Ref. [3] that general workers on the mine who are working in the high background radiation area receive an incremental effective dose of 0.24 mSv/a from external exposure, assuming eight working hours per day and 251 days per year. For workers involved in mining, including the trucking of ores, the corresponding incremental dose from external exposure is more than 1.0 mSv/a, while those working in the disposal sites receive an incremental dose of 0.7 mSv/a from external exposure and 2.38 mSv/a from internal exposure to ^{220}Rn.

(b) Public exposure: The average radiation level in the urban area of about 4.3 km^2 is 0.121 μGy/h, which is 30% higher than the indoor level of 0.0922 μGy/h [13]. The additional effective dose received from external exposure for members of the public in the urban area is 0.044 mSv/a, while the corresponding level for internal exposure to ^{220}Rn is 1.84 mSv/a. These exposures would be higher if inhalation of dust containing thorium series radionuclides from the mining sites and the disposal sites were considered.

4.2. Environmental radiation in the Baotou urban area and surrounding area of the plants

4.2.1. Radiation levels

The data on radiation levels were again obtained by airborne and in-situ gamma spectrometry and dosimetry [3]. The background radiation level in Baotou is 0.065 μGy/h. Higher radiation levels were observed in an area of about 7 km^2, located mostly at the BTISP, including the sorting plant, RE plant and steel refining plant, as well as in the areas where NORM residues are stored. The absorbed dose rates were mostly in the range 0.5–1 μGy/h, the maximum being 1.518 μGy/h. The dose rates in the central area of the RE plants ranged from 0.2 to 0.6 μGy/exceeding 0.6 μGy/h in some plants. Moreover, a number of ‘hot spots’ of radiation levels to some extent were found in some RE plants in the city. Nevertheless, most of these plants are closed. The dose rate around the slag stock is about 0.6–2 μGy/h. Contamination is distributed in the upper 20 cm layer of soil, where the activity concentration of thorium is 0.05–0.35 Bq/g. The radiation level in the plant and its surrounding environment ranged from the background level up to 0.143 μGy/h. The level in the tailings pond without water cover was generally in the range 0.65–1.2 μGy/h, with a maximum of 1.316 μGy/h.

The contaminated area is extended southward more than 2 km by dust flying from the tailings pond; dose rates here were 0.085–0.15 μGy/h. The contaminated topsoil was generally in the range 0.08–0.2 Bq/g, but more than 0.4 Bq/g near the tailings pond.

From the previous monitoring report [13], the aerosol measurements of ^{220}Rn progeny varied between 75.4 and 590.1 MeV/L (0.012–0.095 μJ/m^3), and the average was 243 MeV/L (0.038 μJ/m^3) in areas such as the steel refinery plant, RE plants, tailings pond and blast-furnace slag dump. In the urban area, the PAEC of ^{220}Rn progeny was in the range of 68.9–229.1 MeV/L (0.010–0.037 μJ/m^3), with an average of 136.5MeV/L (0.022 μJ/m^3).

The present PAEC of ^{220}Rn progeny is 25–500 MeV/L (0.004–0.080 μJ/m^3) in the environment of an RE plant, with an average of 126 MeV/L (0.020 μJ/m^3). In the Baotou City area, the PAEC is in the range 69–125.6 MeV/L (0.011–0.020 μJ/m^3), with an average of 94 MeV/L (0.015 μJ/m^3) [5].

The absorbed dose rates are observed to be 0.300, 0.328 and 0.257 μGy/h in the sorting workshop, the sintering workshop and the RE workshop, respectively.

4.2.2. Exposures

(a) Occupational exposure: The additional effective doses from external exposures for workers in the sorting workshop, the sintering workshop and the RE workshop are 0.36, 0.41 and 0.29 mSv/a, respectively. The corresponding value for those working in the ferrous slag dump is 0.33 mSv/a, assuming 2 working hours per day. For those working in the slag brick plants, their additional effective dose from external exposure is 0.61 mSv/a, assuming 8 hours per day, while for those working in the tailings pond area the additional dose is 0.28 mSv/a assuming 2 working hours per day. The contribution of ^{220}Rn progeny to the effective dose from internal exposure in the above mentioned areas is 1.05–5.27 mSv/a based on the aerosol measurements from the previous monitoring reports [13].

(b) Public exposure: The additional effective dose from external exposure is 0.043 mSv/a for members of the public living in the vicinity of the southern tailings pond and is thus not significant. However, attention must be drawn to airborne dust, which forms an aerosol of fine particles dispersed from the tailings pond. Inhalation of these particles will cause an increase in internal exposure. The indoor effective dose for the normal buildings containing no slag is 1.86 mSv/a in the Baotou City area, but the dose exceeds 2.0 mSv/a for most of the buildings made of slag bricks. Some of these buildings are observed to have an internal exposure from indoor ^{220}Rn progeny three times higher than that in the normal buildings. Based on the aerosol measurements in the urban area, the internal exposure from outdoor ^{220}Rn progeny varies from the previously determined value of 1.26 mSv/a [13] to the recently-determined value of 0.02 mSv/a [5].

5. CONCLUSIONS AND DISCUSSION

5.1. Conclusions

5.1.1. Occupational exposures and public exposures

The additional effective dose from external exposure for workers in the plants ranges from 0.29 to 0.41 mSv/a, while for those working in mining areas it varies from 0.24 to 1.0 mSv/a. If the inhalation of aerosol and dust containing thorium is considered, the additional effective dose for many workers probably exceeds 1.0 mSv/a.

The environmental radiation level around the plants is nearly the same as the background radiation level. The additional exposure received by members of the public is not significant. However, the indoor radiation level is higher in the buildings using slag materials; hence, the effective dose increases.

5.1.2. Use and management of NORM residues

Tailings and RE slag are stored as mineral reserves. A small portion of waste rock and some ferrous slag are converted into construction materials. Under the radiation safety regulations of management, waste iron has been recovered from blast furnace slag as well as raw materials for cement and bricks that meet the building material radiation safety requirements. The development and exploitation of NORM residues is of importance for waste minimization. The improvement of the off-gas cleaning technique and the application of acid recovery have resulted in lesser discharges of exhaust gas and liquid effluent, and so the environmental quality in Baotou has improved.

Local government closed those RE plants that did not meet the requirements of environmental discharges, in order to reduce the number of RE plants. The RE plants that are already located in the urban area have been requested to move to the designated industrial park, to improve their technique and to raise production capability, as well as to submit to the supervision of the Environmental Department.

In order to reduce airborne silt and dust, water spraying is applied regularly in the mining area to prevent dust from becoming airborne. While the tailings pond is immersed with incoming liquid effluent, water spraying is also applied. In addition, trees have been planted in the surrounding area of the tailings pond, and the effect is quite good.

The Environmental Department regularly supervises the working site and its surrounding area to ensure the safety of the workers and control of waste discharges. The radioactive safety management of the production of RE raw materials, the discharge of exhaust gas and other effluents and commercial use of RE slag should be strengthened.

The government is concerning itself with plans to resolve the problems of contaminated soil and the potential risk posed by the facilities. A new sorting plant in Bayan Obo started to supply water and refine slag simultaneously in 2009. The annual load is 5.5 million t. All tailings are disposed of in the tailings pond at the Bayan Obo site. The Baogang tailings pond will be closed gradually; it is expected that the pollution problems of the tailings ponds in Baotou will be solved in the near future.

Radiation safety training courses offered by the Environmental Department are conducted regularly for employees in the production and management departments to raise their knowledge of both safety and radiation protection.

5.2. Discussion

The development and exploitation of residues results in the dispersion of materials or products with elevated radioactivity and the collective dose increases significantly. A typical example of the spread of NORM is the use of residues to make building bricks. Therefore, the ALARA principle should be fully taken into account in studying the exploitation of the residues. A criterion should be set to define the uses of NORM residues within the categories of ‘banned’, ‘conditional use’ and ‘unconditional use’ in terms of activity concentration of NORM residues. Meanwhile, the uses of construction materials made of NORM residues should be restricted.

Remediation or decommissioning of the closed RE plants should be considered. All RE slag or other NORM residues, the contaminated soil or materials left in the plants and their surroundings need cleaning or proper treatment and disposal.

Workers in non-uranium mining and processing are considered to be members of the public according to radiation protection regulations in China, but many of them receive more than 250 μSv/a of additional individual exposure, even as high as the 1 mSv/a dose limit for the public. Therefore, they should adopt an effective radiation protection programme.

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NORM USED IN FRENCH INDUSTRIAL FACILITIES: URANIUM AND THORIUM SERIES ACTIVITY CONCENTRATIONS AND ASSOCIATED OCCUPATIONAL EXPOSURES

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Abstract

According to Council Directive 96/29 Euratom, handling or storage of NORM has to be considered from the radiological protection point of view. This directive has been implemented in French regulations by, in particular, the Ministerial Order of 25 May 2005 related to activities involving NORM that is not used for its radioactive properties. It imposes a requirement for radiological characterization of materials and for assessments of the effective dose received by workers. ASN and IRSN have already reviewed 90 studies which provide information about activity concentrations of materials and occupational exposure of nine different types of industrial facility. These data show that activity concentrations vary considerably, according to the type of material and industrial activity. Waste generally contains the highest activity concentration of radionuclides of natural origin. Activity concentrations in raw materials and products sometimes exceed the criterion recommended by the IAEA for the regulation of NORM. The activity concentrations of ^{226}Ra and its daughters in waste or of ^{210}Pb and its daughters in ash and dust related to high temperature processes are sometimes greater than the activity concentrations of the other radionuclides of the ^{238}U series. Concerning occupational exposure, doses reported by operators range from <1 to 82 mSv/a. Based on the feedback from French industries, the following conclusions can be drawn: assessments are still expected for some industries; about 15% of calculated doses are greater than the effective dose limit of 1 mSv/a for the public and need further examination; the highest doses correspond to the production of thorium compounds; external and internal exposure are often of the same order of magnitude; some types of industrial facility currently not covered by the French ministerial order, such as paper mills, are affected by the NORM issue.

1. INTRODUCTION

Some industrial activities such as ceramic production or coal combustion in thermal power stations involve the use of NORM (material usually regarded as non-radioactive but containing radionuclides of natural origin). Handling or storage of this material can lead to a significant increase in occupational exposure. According to Council Directive 96/29 Euratom [1], this matter has to be considered from the radiation protection point of view. French authorities implemented this directive to the French health and labour codes detailed in the Ministerial Order of May 25, 2005 [2]. It sets out a list of ten types of industrial activities concerned with NORM and operators have to assess the effective doses received by workers on the basis of a radiological characterization of raw materials, by-products and waste involved in or produced by the technological processes. ASN asked IRSN to evaluate the methods adopted by some operators for assessing these doses and to draw the first conclusions in terms of radiation protection. Based on data presented by operators in the 90 studies received so far, a summary of the results of these studies in terms of activity concentrations and occupational exposure is presented.

2. INVENTORY OF DATA PRESENTED IN THE STUDIES RECEIVED

Since the publication of the Ministerial Order of May 25, 2005 [2], ASN and IRSN have already received 90 studies which provide activity concentrations of materials and doses calculated for nine types of industrial activities (see Table 1). Most of the studies deal with occupational exposure and about 91% of them report on effective doses received by workers.

Of the 90 studies, 43 present the results of activity concentration measurements carried out mainly by gamma spectrometry for about 500 different samples. These represent more than 4200 measurement results. Radionuclides of the ^{238}U, ^{232}Th and ^{235}U series and ^{40}K represent 47%, 29%, 15% and 9% of the measurements, respectively. The proportions of activity concentrations above the detection limit (LD) were 69% for the ^{238}U series or ^{232}Th series, 32% for the ^{235}U series and 59% for ^{40}K.

TABLE 1. ANALYSIS OF STUDIES, BY TYPE OF INDUSTRIAL ACTIVITY

Industrial activity	Proportion of total (%)
Production of refractory ceramics and the smelting, metallurgy and glass industries using them	49
Coal combustion in power plants	16
Production of zircon and zirconia, and the smelting or metallurgy plants using them	15
Processing of tin, aluminium, copper, titanium, niobium, bismuth and thorium ores	7
Production of phosphate fertilizers and phosphoric acid	6
Production or use of thorium compounds	3
Separation and purification of rare earths and the production of pigments containing them	2
Spas	1
Underground water treatment by filtration	1
Production of titanium dioxide	0

3. URANIUM AND THORIUM SERIES ACTIVITY CONCENTRATIONS

3.1. Measurements taken into account

The activity concentrations measured mainly by gamma spectrometry were analysed at IRSN. The uranium and thorium series were divided into groups of radionuclides as recommended by the European Commission [3]. For each chain segment, an activity concentration was determined by the following method [4]. For a given sample, if only one radionuclide activity concentration of a group is measured, this activity concentration stands for the group. If several radionuclide activities are measured and their measurements are consistent, the average of the more reliable activity concentrations stands for the group. For example, regarding the chain segment of ^{226}Ra+,[1] deconvolution of the ^{226}Ra gamma ray (186.2 keV) and the ^{235}U gamma ray (185.7 keV) cannot be done accurately by the classic HP-Ge detectors used mainly in the studies (detectors made of high purity

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[1] The symbol ‘+’ after a radionuclide denotes a segment chain headed by that radionuclide. For example, ^{226}Ra+ corresponds to the following chain segment: ^{226}Ra, ^{222}Rn, ^{218}Po, ^{218}At, ^{214}Pb, ^{214}Bi and ^{214}Po.

germanium), whereas the activity concentration of ^{214}Pb or ^{214}Bi is easy to measure with those detectors. So the average of the activity concentrations of ^{214}Pb and ^{214}Bi stands for the ^{226}Ra+ group.

3.2. Activity concentrations and disequilibrium in NORM in French industries

As a first step, all samples presented by the operators were classified into four categories: raw materials, products, waste and environment. The category ‘environment’ represents a fifth of the samples and includes all the samples collected in the environment, such as soil, sediments and vegetables. These data have been excluded from our work. The category ‘products’ includes final products and by-products and the category ‘waste’ includes solid waste, effluents, sludge and dust. Of the materials considered by the operators, 26% are raw materials, 34% are products and 39% are waste.

In the second step, twelve industrial activities were identified: coal combustion, the glass industry, the production and use of zircon and zirconia, the production of phosphate fertilizers, the production of refractory ceramics, the production and use of thorium compounds, spas, the processing of aluminium ores and kaolin ores, the separation and purification of rare earths, the processing of titanium ores and underground water treatment by filtration. All the samples were classified according to these categories.

Eventually, for each category of materials and for each chain segment, the minimum, maximum and median values, as well as the first and third quartile values were determined. Figure 1 shows the activity concentrations of the chain segments measured most often, ^{226}Ra+ and ^{228}Ra+.

3.3. Conclusions relating to activity concentrations

Based on the feedback from French industries, the following conclusions can be drawn:

(a) The activity concentrations vary considerably according to the type of material and industrial activity;
(b) Waste generally contains the highest activity concentration of radionuclides of natural origin;
(c) Of the materials with activity concentrations higher than 1000 Bq/kg (the IAEA’s criterion for the regulation of NORM), 55% are waste, 29% are products and only 16% are raw materials.

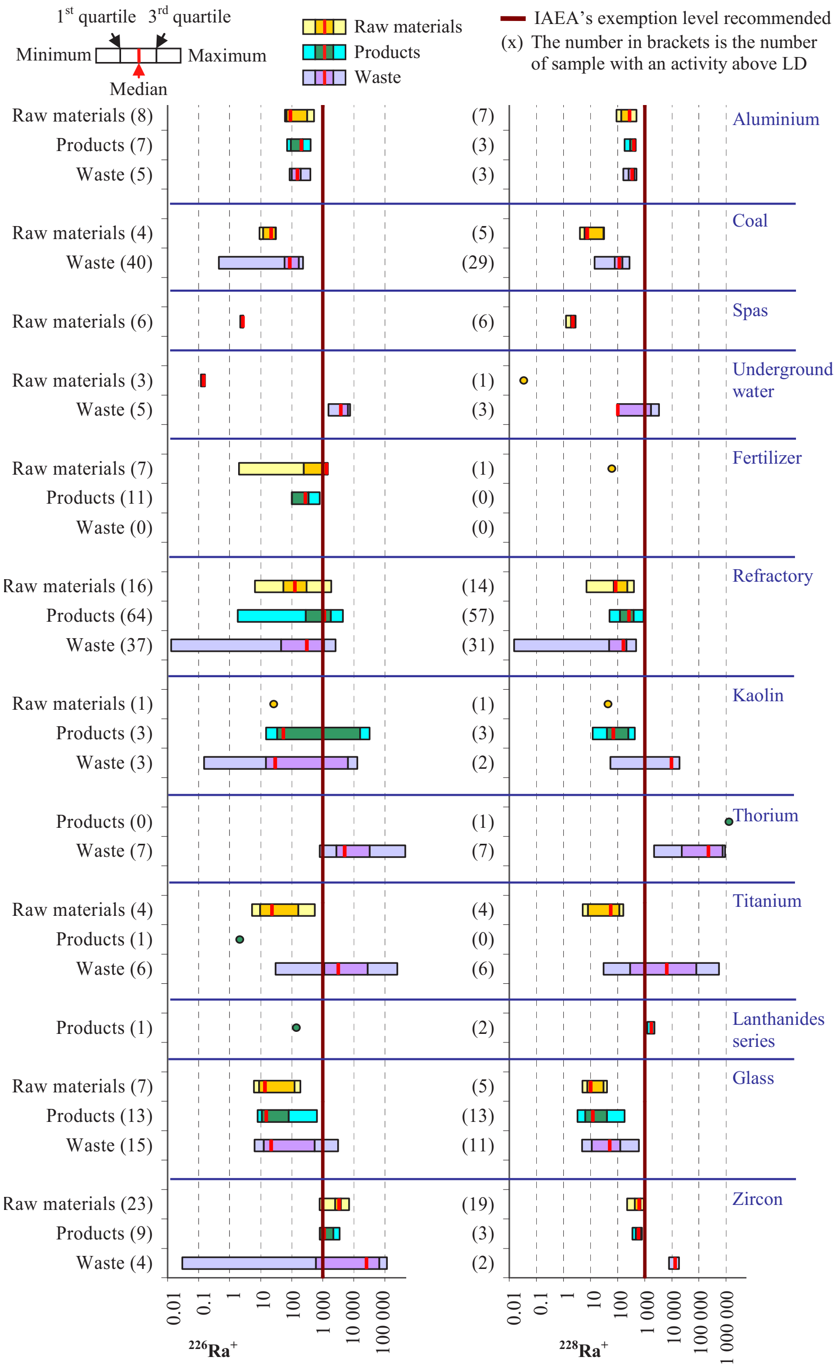

FIG. 1. Activity concentrations of ^{226}Ra+and ^{228}Ra+ (Bq/kg).

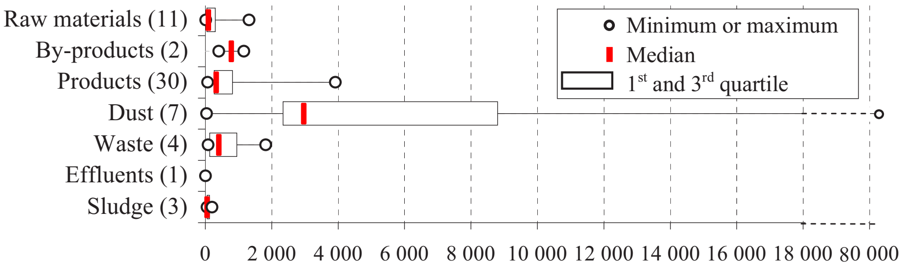

FIG. 2. Activity conentrations of ^{210}*Pb+ in the refractory industries (Bq/kg).*

(d) The number of radionuclides measured and the accuracy of their activity determination are barely enough to assess the secular equilibrium or radioactive disequilibrium that occurs in NORM.

(e) Inbalances have been identified: the activity concentration of the ^{226}Ra+ group in waste (such as the category ‘underground water’) or of the ^{210}Pb+ group in ash and dust related to high temperature processes is greater than those of the other groups of the ^{238}U series. For example, Fig. 2 illustrates an excess of ^{210}Pb in dust from the refractory industries.

4. OCCUPATIONAL EXPOSURES

4.1. Data collected

The Ministerial Order of May 25, 2005 requires operators to assess the occupational effective doses and to take into account external exposure, as well as internal exposure by inhalation of dust and radon progeny. Accordingly, operators have assessed occupational exposures for more than 400 workplaces. A third of the doses are less than 0.1 mSv/a, half are less than 0.25 mSv/a, while 15% of the doses are greater than the effective dose limit of 1 mSv/a for the public in France [5] and so need further examination.

Firstly, some doses calculated by operators do not take account of external exposure or internal exposure by dust inhalation even if this route could be a significant pathway of exposure. Secondly, only some of the doses consider also the exposure due to inhalation of radon progeny. Thirdly, only some operators take into account the exposure due to the natural background in their assessment. Finally, the doses received by workers vary considerably according to the type of industrial facility. Owing to the differences in approach adopted by operators, it is not possible to compare directly the doses reported. On the one hand, when data are available, the exposure due to natural background was subtracted from the

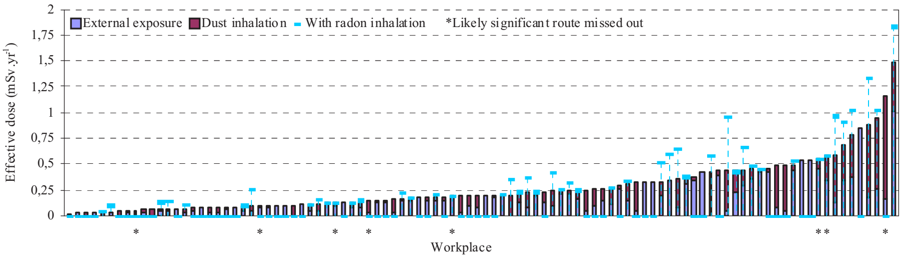

FIG. 3. Production and use of refractory ceramics — incremental effective dose

effective doses reported by operators. On the other hand, in order to compare the effective dose for each type of industrial activity and to identify the significant route of exposure, IRSN has analysed the doses calculated for workers due to external and internal exposure for each type of industrial activity. Data relative to exposure to radon have been considered specifically.

4.2. Incremental effective doses above the natural background

4.2.1. Production of refractory ceramics and the smelting, metallurgy and glass industries using them

The production of refractory ceramics and the smelting, metallurgy and glass industries using them is the industrial activity assessed most often: more than 100 workplaces have been assessed. Figure 3 shows the incremental effective dose. For two workplaces, the doses are greater than 1 mSv/a with a maximum of 1.5 mSv/a, consistent with the literature [6].

4.2.2. Production of zircon and zirconia and the smelting or metallurgy plants using them

The production of zircon and zirconia and the smelting and metallurgy plants using them is the industrial activity assessed second most often: more than 60 workplaces have been assessed. Figure 4 shows the incremental effective dose reported by the operators. For eight workplaces, the dose is greater than 1 mSv/a with a maximum dose of 2.3 mSv/a. This result is consistent with those published in the literature for zirconia production by the thermal process [6]. Moreover, it is worth mentioning that the highest doses correspond to two studies in which a hypothetical maximum exposure period of 1600 h/a has been considered.

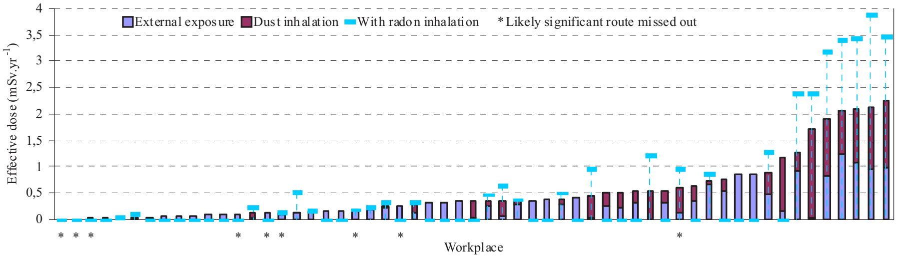

FIG. 4. Production and use of zircon and zirconia — incremental effective dose.

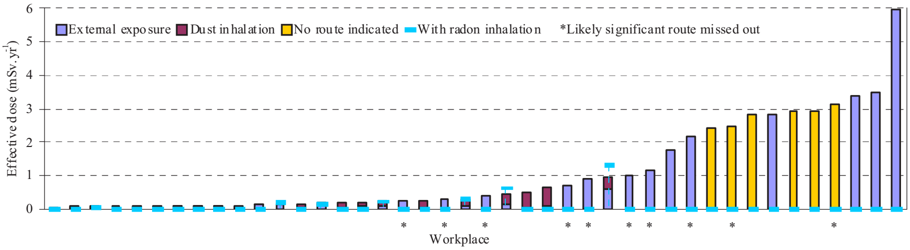

FIG. 5. Processing of ores (Sn, Al, Ti, Nb) — incremental effective dose.

4.2.3. Processing of tin, aluminium, titanium and niobium ores

The processing of tin, aluminium, titanium and niobium ores is the industrial activity assessed third most often: more than 40 workplaces have been assessed. Figure 5 shows the incremental effective dose. For 13 workplaces, the doses are greater than 1 mSv/a with a maximum of 6 mSv/a and are consistent with results reported in the literature [6, 8]. It is worth mentioning that the highest doses correspond to only one study. New measurements in 2008 show an important reduction in dose rate: based on these new measurements, the maximum dose decreases to 3.2 mSv/a.

4.2.4. Coal combustion in power plants

Thirty workplaces for coal combustion in thermal power plants have been assessed. Figure 6 shows the incremental effective dose reported by the operators. For each workplace, the dose is less than 1 mSv/a with a maximum dose of 0.4 mSv/a, which is consistent with doses reported in the literature [7].

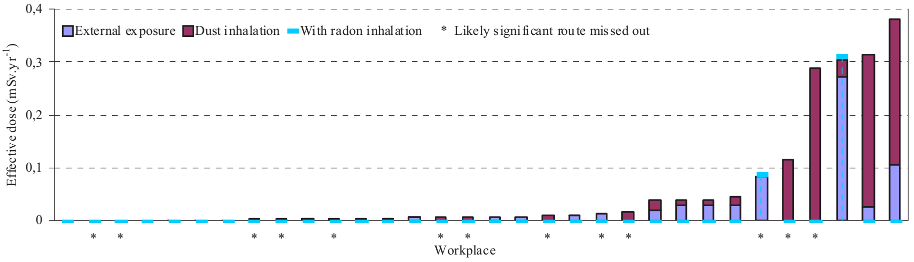

FIG. 6. *Coal combustion — incremental effective dose.*

4.2.5. Other industrial activities

For the production or use of thorium compounds, six workplaces have been assessed. For two workplaces, the doses are greater than 1 mSv/a with a maximum dose of 82 mSv/a. This dose is mainly due to dust inhalation. In order to reduce occupational exposure, the operator was intending to equip workers with personal protective equipment, clean the workplaces periodically and install air filtration. These actions should significantly reduce the doses but IRSN and the French authorities have not yet received the new study.

For the production of phosphate fertilizers, six workplaces have been assessed. All doses are less than 1 mSv/a with a maximum dose of 0.5 mSv/a, consistent with doses reported in the literature [6, 9].

For the separation and purification of rare earths, three workplaces have been assessed. All doses are less than 1 mSv/a with a maximum dose of 0.3 mSv/a, consistent with values reported in the literature [9].

4.3. Exposure pathways

Workplaces have been studied sufficiently to enable the significant route of exposure to be identified for only four types of industrial activities. Figure 7 shows the contributions of external and internal exposure for these four types of industrial activity.

(a) For coal combustion, the data show clearly that external exposure is the significant route of exposure;

(b) For the production of refractory ceramics and the smelting, metallurgy and glass industries using them, and for the production of zircon and zirconia and the smelting or metallurgy plants using them, the data collected do not clearly show any dominant route of exposure, although external exposure seems to be the most important pathway.

(c) For processing of ores, no dominant route of exposure could be identified.

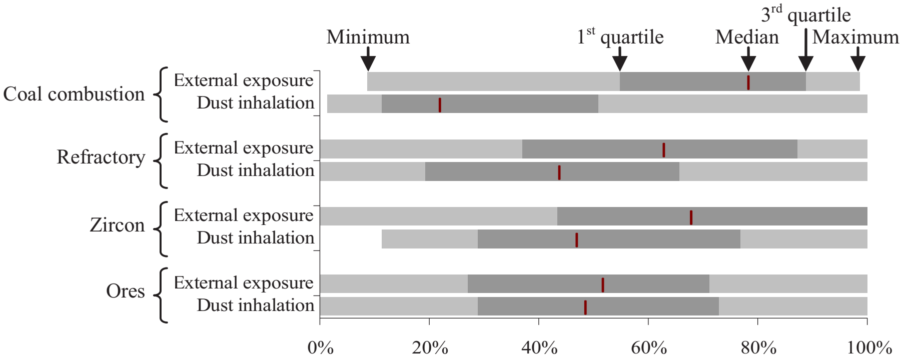

FIG. 7. Contributions of external exposure and dust inhalation.

5. CONCLUSIONS

Based on the feedback gained by IRSN and French authorities from the studies conducted by operators, the following conclusions can be drawn, in addition to those presented in Ref. [4]:

(a) Assessments are still expected for some industrial activities referred to in the Ministerial Order of May 25, 2005 [2]. For example, in the near future, studies dealing with occupational exposure due to underground water treatment by filtration should be carried out.

(b) About 15% of the calculated doses are greater than 1 mSv/a and need further examination.

(c) The highest doses were found in facilities that produce materials involving thorium (maximum 82 mSv/a) and that process tin, aluminium, titanium and niobium ores (maximum 6 mSv/a). For the other types of industrial activity, the maximum dose remained below a few millisieverts per year.

(d) The doses due to external and internal exposure are often of the same order of magnitude.

(e) Some types of industrial facilities currently not referred to in the French ministerial order, such as paper mills, are affected by the NORM issue and could be added to the list of industrial facilities established by the Ministerial Order of May 25, 2005 [2].

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ECOLOGICAL AND RADIOLOGICAL CONSEQUENCES OF A HALF CENTURY OF OPERATION OF A MONAZITE STORAGE FACILITY

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Abstract

Since 1960 more than 82 000 t of monazite concentrate have been stored at the storage facility situated in the south-eastern part of the Sverdlovsk region (Russian Federation). The complex assessment of the radiation situation on-site and in the vicinity of the monazite storage facility was conducted in 1995–2009. It was shown that there is no elevation of ^{232}Th concentration in the 0–5 cm soil layer in comparison with the 5–10 cm layer. Soil contamination by ^{232}Th in the range of 27–70 Bq/kg was found only at a few points. It was concluded that the contamination spots were due to occasional spills during loading and repackaging operations. The average ^{220}Rn concentration in the atmosphere inside the warehouses is 3800 Bq/m^3 (range 300–14 000 Bq/m^3). In the outside atmosphere near the warehouses the average thoron concentrations are 470 Bq/m^3 in winter and 760 Bq/m^3 in summer. Four modes of ^{212}Pb thoron progeny aerosols with different size distributions were evident from the measurements: an unattached fraction, with an activity median thermodynamic diameter (AMTD) of 2 nm (27.5 ± 13.0%); a nucleation mode, with an AMTD of 8 nm (2.5±0.6%); an Aitken mode, with an AMTD of 18 nm (2.0 ± 0.6%) and an accumulation mode, with an activity median aerodynamic diameter (AMAD) of about 300 nm (68 ± 14%). Values of dose conversion factor (DCF) between thoron equivalent equilibrium concentration exposure and effective dose were calculated, taking into account both the measured radioactive aerosol size distribution and the dissolution rate of real ^{212}Pb aerosols in imitation body fluids determined in specially conducted experiments. It was shown that the nominal value of the DCF is in the range 80–90 nSv per Bq·h·m^{-3}. It is demonstrated that outside the controlled zone of the storage facility there is no excess radiation influence on the environment or on the human population.

1. INTRODUCTION

The history of the monazite storage facility in the south-eastern part of the Sverdlovsk region (Russian Federation) began on 10 January 1960 when the first railroad train was discharged and boxes containing paper bags filled with 50 kg of monazite sand were placed in the former granary barns of the Federal Reserve.

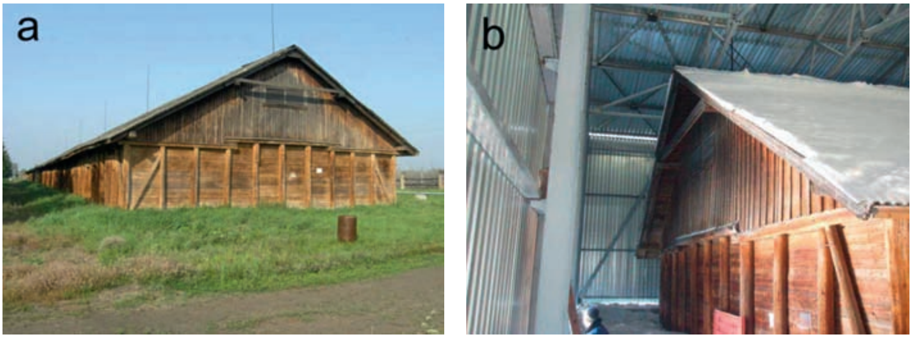

FIG. 1. One of the former granaries of the monazite storage facility before and after the repair work.

Monazite contains 5–10% ThO_2 (sometimes up to 30%) and about 0.2 % U_3O_8. Since 1960, more than 82 000 t of monazite concentrate have been stored at this facility in 19 wooden warehouses (former granaries) and 4 hangars. The granary warehouses were built in 1940 and were not adapted for the storage of high (up to 4 m) stacks of heavy boxes. Most of the warehouses were in need of serious repair at the end of the twentieth century (Fig. 1(a)). As a result, high levels of concern by the local authorities and the general public about the possible influence of the monazite storage facility on the ecological situation and the health of people existed from 1985 to 1990. During recent years, considerable work on repairing the old granaries was conducted. The new hangars were built above the old storage buildings (Fig. 1(b)) and the probability of accidental monazite contamination was minimized.

Many and various pseudoscientific speculations arose about a connection between the radioactive material in the granaries and different kinds of oncological and non-oncological diseases among the local population and other people working near the storage facility. To obtain the answer on the real levels of radiological influence on the facility workers, population and environment, a special complex assessment of the radiation situation on-site and in the vicinity of the monazite storage facility was conducted in 1995–2009.

2. METHODS AND INSTRUMENTATION

Measurements of ambient gamma radiation dose rate were conducted by standard equipment. The specific activity of radionuclides in soil and plants was measured by spectrometers equipped with either high purity germanium or NaI(Tl) detectors.

LR-115 track detectors of the types used most widely in Russia were calibrated for sensitivity to radon and thoron [1]. The results of the calibration are given in Table 1. To have the possibility of accounting for the thoron concentration in the ambient air during the field measurements, thoron-sensitive detectors developed at the Institute of Radiation Hygiene (St. Petersburg) were modified. The diameter of the holes for radon penetration into the detector was considerably increased and the silicone anti-thoron membrane was replaced by filter material for preventing the entry of radon and thoron progeny into the detector volume. The parameters of the modified detectors are also shown in Table 1.

The modified track detectors in combination with the detectors REI-4 developed by the Radioecology Studies company (Moscow) were used for radon and thoron concentration measurements in the monazite storage facility. The aerosol size distribution measurements were conducted by the combined use of a diffusion wire screen battery and a cascade impactor. For the aerosol distribution measurements in the AMTD range from 1 to 50 nm, a special wire-screen diffusion battery was designed. For better separation of different aerosol modes and to minimize systematic and random errors, individual alpha activity measurements were conducted on every wire screen after attaining equilibrium between ^{212}Pb and ^{212}Bi in the diffusion battery elements. The cascade impactor enabled the radioactive aerosol size distribution in the range of 0.3–10 μm to be estimated.

3. MONITORING RESULTS

3.1. Direct assessment of monazite dust emission from the storage building

To estimate the amount of radioactive dust emitted from the ventilation windows, a dust filter was installed on one of the open dormer windows. It should be noted that under usual conditions the dormer windows in all facilities are always closed. The filters were exposed during the summer season for three years. The exposure periods were 30, 76 and 130 d, respectively. After exposure, the filters acquired a gray colour owing to fine aerosol emissions of organic origin (old and new birds' nests were found inside the storage buildings). Immediately after removal of the filters it was possible to detect the activity of the ^{232}Th decay products ^{212}Pb and ^{212}Bi. After one week, the activity on the filters was in all cases less than the detection limit (0.2 Bq of ^{232}Th in equilibrium with its decay products). This showed that even with open ventilation windows the annual airborne emission of monazite was less than 2.5 mg.

TABLE 1. SENSITIVITY TO RADON AND THORON OF LR-115 TRACK DETECTORS USED IN THE RUSSIAN FEDERATION

Track detector type	Thoron detection efficiency (track/cm^2 per Bq·m^{-3}·d)	Radon detection efficiency (track/cm^2 per Bq·m^{-3}·d)	Thoron/radon sensitivity ratio
Detector IRR-3m of the Radioecology Studies company (Moscow)	3.77×10^{-4}	3.79×10^{-2}	0.0099
Detector REI-4 of the Radioecology Studies company (Moscow)	2.38×10^{-4}	1.94×10^{-2}	0.012
Detector of the Institute of Radiation Hygiene (St. Petersburg), initial configuration	1.60×10^{-3}	2.24×10^{-2}	0.071
Detector of the Institute of Radiation Hygiene (St. Petersburg), modified configuration IIE UB RAS	9.69×10^{-3}	5.2×10^{-2}	0.19
Detector of the Radium Institute (St. Petersburg)	9.97×10^{-4}	2.97×10^{-2}	0.034

The total fallout of monazite dust on-site was assessed by meteorological plane-tables covered by filter material. The filters were exposed simultaneously with the filters on the dormer windows over a period of 30–137 d. The thorium fallout in the close vicinity of the warehouses during the exposure period was less than the detection limit. This means that the thorium deposition rate due to aerial transfer on the site of the monazite storage facility was less than 0.15 Bq/m^2 per month, or less than 100 g of monazite on the whole site during its 50 year history. These results are in accordance with expectations because, for a typical monazite grain size of about 0.12 mm (range 0.08–0.5 mm) at a relative density of 4.9–5.5, any migration of monazite particles out of the warehouses by weak airflows would be highly improbable.

3.2. Measurement of radionuclide activity concentrations in the soil

Radionuclide activity concentration measurements were made on 34 samples taken on-site and four samples taken off-site. The sampling depth was 7 cm including the sod cover. At six points on the site, soil specimens were

TABLE 2. RADIOACTIVITY IN TOPSOIL AT THE SITE OF THE STORAGE FACILITY

	Activity concentration at a depth of 7 cm (Bq/g)			
	Average	Standard deviation	Minimum	Maximum
^{232}Th	0.026	0.018	0.012	0.091
^{226}Ra	0.052	0.0091	0.029	0.068
^{40}K	0.344	0.0	0.269	0.415
^{137}Cs	0.019	0.0077	0.0063	0.041

sampled from layers at depths of 0–5, 5–10, 10–15, and 15–20 cm. The measurement results are shown in Tables 2 and 3. The concentrations of ^{232}Th and ^{226}Ra in the soil on the site correspond to the average concentrations of radionuclides of natural origin for soils in this location. The distribution of ^{137}Cs concentrations in the soil from global and Chernobyl fallout showed that there had been no mechanical disturbance of the soil during the last ten years. There was no elevated ^{232}Th concentration in the 0–5 cm soil layer in comparison with the 5–10 cm layer. This supports the conclusion that there was no significant monazite dust emission from the storage buildings.

Soil ccontamination by ^{232}Th in the range 0.027–0.070 Bq/g above the average level was found only at a few points. The contaminated points are distributed randomly over the site. By radionuclide ratio analysis it was proved that such elevated concentrations are statistically significantly different from the average level [2]. It was proved that the local contamination spots were developed due to occasional spills during loading and repackaging operations. The total mass of monazite spills over the storage facility site is estimated to be about 500 kg.

3.3. Radionuclide concentrations in plants and mushrooms

Typical species of plants growing on the site of the storage facility were sampled. After ashing, the activity concentrations of radionuclides of natural origin were measured by a germanium detector. In all plant samples, ^{226}Ra, ^{232}Th and ^{137}Cs (due to global and Chernobyl fallouts) were not detected — only ^{40}K and cosmogeneous ^{7}Be were detected.

Two species of mushrooms were found on the site of the monazite storage facility. The radionuclide activity concentrations measured in the mushrooms are shown in Table 4. No activity of ^{226}Ra and ^{232}Th was found in the mushrooms.

TABLE 3. RADIOACTIVITY IN VARIOUS LAYERS OF SOIL AT THE SITE OF THE STORAGE FACILITY

	Depth of layer (cm)	Activity concentration (Bq/g)			
		Average	Standard deviation	Minimum	Maximum
^{137}Cs	0–5	0.0188	0.0096	<0.002	0.0315
	5–10	0.0133	0.0065	<0.002	0.0227
	10–15	0.0076	0.0058	<0.002	0.0177
	15–20	0.0036	0.0052	<0.002	0.0120
^{226}Ra	0–5	0.0158	0.0041	0.009	0.0204
	5–10	0.0141	0.0030	0.009	0.0182
	10–15	0.0147	0.0023	0.011	0.0170
	15–20	0.0171	0.0053	0.0119	0.0290
^{232}Th	0–5	0.0224	0.0105	0.0151	0.0480
	5–10	0.0196	0.0062	0.0112	0.0330
	10–15	0.0195	0.0042	0.0140	0.0276
	15–20	0.0194	0.0041	0.0142	0.0253
^{40}K	0–5	0.357	0.0295	0.309	0.396
	5–10	0.348	0.0518	0.261	0.405
	10–15	0.373	0.0318	0.346	0.446
	15–20	0.358	0.0383	0.271	0.402

The ^{137}Cs plant–soil distribution coefficients of 0.2 for *Leccinum scabrum* and 8.7 for *Suillus granulatus* are typical for the mushrooms concerned.

3.4. Concentrations of radon, thoron and their progeny in air

Measurements of ^{220}Rn and ^{222}Rn concentrations were made in both summer and winter using nuclear track detectors. The average ^{220}Rn concentration in the atmosphere inside the warehouses was 3800 Bq/m^3 (range 300–14 000 Bq/m^3). There was practically no difference between the average summer and winter concentrations. In the outside atmosphere near the walls of the warehouses the average thoron concentrations were 470 Bq/m^3 in winter and 760 Bq/m^3 in summer. Near an open dormer window during the summer period the ^{220}Rn concentration was 670 Bq/m^3. The average thoron concentration at a

TABLE 4. RADIOACTIVTY IN MUSHROOMS AT THE SITE OF THE STORAGE FACILITY

	Activity concentration (Bq/g)			
	Non-treated samples		Air dried samples	
	^{137}Cs	^{40}K	^{137}Cs	^{40}K
Leccinum scabrum	0.0005	0.150	0.0035	1.10
Suillus granulatus	0.0199	0.105	0.236	1.24

distance of about 1 m from the wall of the warehouse decreased to 230 Bq/m^3. On the surrounding site, the average outdoor ^{220}Rn concentration was about 70 Bq/m^3, but individual values were strongly dependent on the measurement location (the range was 11–150 Bq/m^3). The average ^{222}Rn concentrations inside the warehouses were 220 Bq/m^3 in both summer and winter. The average ^{222}Rn concentrations near the outside walls were 80 Bq/m^3 in winter and 160 Bq/m^3 in summer. On the surrounding site, the average ^{222}Rn concentration was 19 Bq/m^3.

The average thoron equivalent equilibrium concentration (EEC) inside the warehouses was 350 Bq/m^3. It was shown that the instantaneous thoron EEC value in the warehouse could vary by 2–4 times during a 24 h period and up to 10 times during longer periods. On the surrounding site, the thoron EEC values were 15–50 Bq/m^3 near the outside wall; 6–18 Bq/m^3 at 10 m from the warehouse and about 2 Bq/m^3 at 20 m from the warehouse. The thoron EEC values both inside the warehouse and on the surrounding site were strongly dependent on weather conditions, temperature, wind speed and direction.

3.5. Aerosol size distribution of thoron progeny

Four modes of thoron progeny (^{212}Pb) aerosols with different size distributions were evident from the measurements. These modes were identified as the unattached fraction, with an AMTD of 2 nm; a nucleation mode, with an AMTD of 8 nm; an Aitken mode, with an AMTD of 18 nm and an accumulation mode, with an AMAD of about 300 nm. The ^{212}Pb activity concentration was found to be distributed across the four aerosol modes as follows: unattached fraction 27.5 ± 13.0%, nucleation mode 2.5 ± 0.6%, Aitken mode 2.0 ± 0.6% and accumulation mode 68 ± 14% (see Fig. 2).

In work to estimate the lung absorption class of ^{212}Pb aerosols [3], their dissolution from an aerosol filter in imitation body fluids was studied. The filter was placed in Ringer solution at 37°C. Then the filter was rinsed by clean Ringer solution at the same temperature and its activity was determined using a gamma

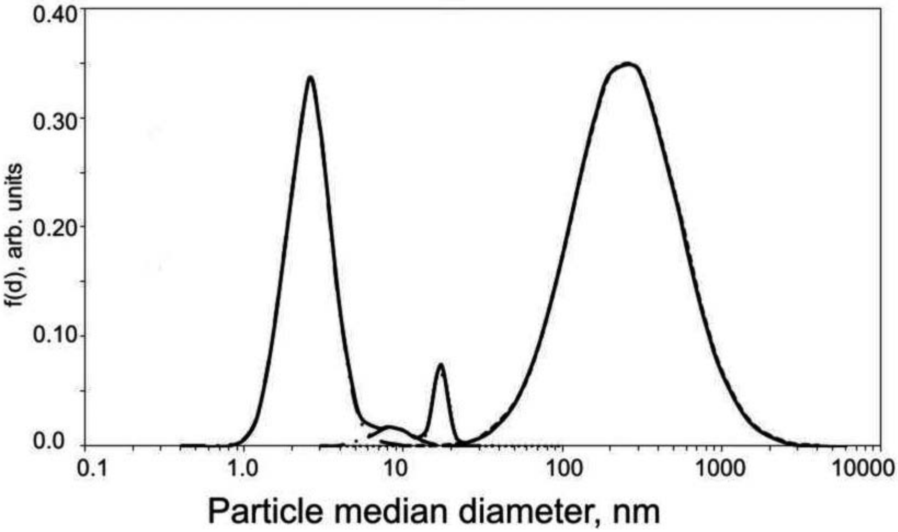

FIG. 2. Aerosol size distribution of ^{212}Pb in the monazite storage facility.

spectrometer. After the activity measurements, the filter was again placed in the solution and the cycles of dissolution and activity measurements were repeated. It is suggested that the kinetics of ^{212}Pb absorption from the respiratory tract to body fluids can be described by the same mechanism as ^{212}Pb dissolution from the aerosol filter to the Ringer solution. For the calculation of the ^{212}Pb dynamics in the respiratory tract, it was assumed that:

$f_r = 0.3$

$\lambda_f = 145\ d^{-1}$

$\lambda_s = 2\ d^{-1}$

where f_r is the fraction of activity with a fast rate of dissolution and λ_f and λ_s are the rates of fast and slow dissolution of ^{212}Pb aerosols, respectively [3]. Pilot experiments to investigate the dissolution rate of aerosols sampled on the filter after the diffusion battery and corresponding to an AMAD of about 300 nm were conducted. A decrease in the fast solubility component for the accumulation mode aerosols was obtained. As a result, it was concluded that the aerosols corresponding to an AMTD of 2–18 nm could be attributed to fast dissolution rate aerosols ($\lambda_f = 145\ d^{-1}$).

The values of dose conversion factor (DCF) between thoron EEC exposure and effective dose were calculated taking into account both the measured radioactive aerosol size distribution and the dissolution rate of real ^{212}Pb aerosols. It was shown that the nominal value of the DCF for light work was 80–90 nSv per Bq·h/m^{-3}, higher than the value of 40 nSv per Bq·h·m^{-3} established by UNSCEAR [4].

The influence of aerosol size distribution on the results of thoron EEC measurements was demonstrated. The permeability coefficient of a standard analytical fibrous filter ranges from 2% for 2 nm AMTD aerosols to 35–40% for aerosols with size distributions in the range of 8 nm AMTD to 300 nm AMAD. It is evident that, for correct interpretation of the monitoring results for thoron progeny, information on the radioactive aerosol size distribution is essential.

3.6. Gamma dose rate

Different zones could be demarcated on the site of the storage facility. Inside the granaries, the average value of ambient dose equivalent rate was 90 ± 20 μSv/h (range 13–160 μSv/h). In the near vicinity of the outside walls, the average dose rate was 44 ± 8 μSv/h (range 5–70 μSv/h). The dose rate between the warehouses was 12 μSv/h. On the perimeter of the monazite storage site, the dose rate varied from 0.1 to 1.0 μSv/h, depending on the distance from the nearest warehouse. In the two nearest settlements, situated 1.0 and 1.5 km from the storage facility, respectively, the gamma dose rate was 0.06–0.12 μSv/h, typical for this region.

3.7. Influence of radiation exposure on the health of monazite storage facility workers

Practically any situation of human exposure to radiation gives rise to various rumours and speculations. The occupational radiation exposure at the monazite storage facility is no exception. In the local ‘green’ mass media, statements such as “*all regular and temporary employees who participated in the loading and unloading operations* [in 1960–1962] *died within two years from unknown causes*” were published.

For the assessment of the radiological consequences of the operation of the monazite storage facility on the basis of archival materials, a register of employees for the period 1960–1997 was created. The total number of workers included in the register is 438. In this register both regular workers and temporary employees who worked on the loading and unloading operations under the contracts were included. For each member of the register, the effective dose and lung equivalent dose were calculated on the basis of individual monitoring data and information about periods of work in the warehouses or on the surrounding site, as logged in the workbooks. The collective effective dose from external radiation for the members of register was 24 man·Sv and the maximum individual dose was 0.33 Sv. The collective lung equivalent dose due to external and internal exposure was 91 Sv and the maximum individual dose was 1.3 Sv. Owing to large uncertainties, the doses obtained during loading and unloading operations were not included.

The cause and age of death were obtained for 101 employees of the storage facility (79 men and 22 women). The main part of this group began work at the storage facility at the beginning of the 1960s. The observed frequency of cancer deaths in the register was compared with the expected frequency obtained by analysis of a database of causes of death for the local population (4679 cases). It was shown that the expected number of cancer deaths for this group was 14, while the observed number was 15. The difference between the expected and observed values is not statistically significant. The collective effective dose for this group of 101 employees was 6.2 Sv. The detriment-adjusted nominal risk coefficient recommended by ICRP is 4.1×10^{-2} Sv^{-1} [5]. So the expected number of radiation induced cancers in this group is less than one case. This estimation is consistent with the observed cancer cases. Thus we can conclude that the radiation impact on personnel, including the operations of loading and unloading of monazite, have not led to a significant impact on their health.

4. CONCLUSIONS

(a) During the half century of operation of the monazite storage facility, no radiation impact on the environment was detected. Elevated levels of ^{220}Rn, ^{222}Rn and their progeny can be detected only within the inner control area of the facility. It has been shown that there was no spontaneous discharge of monazite out of the warehouses due to wind transfer from the dormer windows or other leakage points.

(b) The estimated values of DCF between thoron progeny exposure and effective dose based on the aerosol size distribution measurements and the rate of transfer of ^{212}Pb in body fluids is in the range 80–90 nSv per Bq·h/m^{-3}, which significantly exceeds the value of 40 nSv per Bq·h/m^{-3} established by UNSCEAR.

(c) There is no statistically significant increase of cancer mortality among the former and current workers of the monazite storage facility.

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PERSONAL DOSIMETRY FOR RADIATION WORKERS UPGRADING AN OLD PETROLEUM OIL FIELD

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Abstract

Radiation protection consultations and services were carried out over the last 15 years in Egypt at several petroleum companies. Egypt issued its first NORM Regulation in 2000; this was subsequently modified in 2006. The main aim of the present work was to report dosimetric experience gained during the handling of contaminated equipment in the upgrading of an old oilfield. Optimization of radiation protection was carried out using lead blankets. Film badges were distributed to workers. Films were redistributed to personnel every few weeks. More than 700 film badges were distributed during the working period and workers' health surveillance was carried out, including complete blood analysis. For the 5 month period, individual doses were estimated. The average effective dose was less than 1 mSv.

1. INTRODUCTION

Regionally, NORM activities have been carried out in North Africa and the Middle East since the mid 1990s. NORM studies at Saudi Aramco oilfields in Saudi Arabia were reported in Ref. [1], NORM experience in the Syrian Arab Republic was reported in Ref. [2] and NORM activities in Yemen are reported in Ref. [3]. The atomic energy authorities of several Middle Eastern countries have established regulations concerning NORM, including Egypt [4] and the Syrian Arab Republic [5]. The IAEA has produced publications concerning NORM, including a Safety Report entitled Radiation Protection and the Management of Radioactive Waste in the Oil and Gas Industry [6].

NORM activities in Egypt include the following:

(a) Research and development studies on the treatment of NORM waste using local materials [7];
(b) The regulatory control of NORM contaminated metallic equipment resulting from the Egyptian oil industry;
(c) Participation in AFRA projects dealing with NORM radiation protection consultations and service at various Egyptian oil producing companies;

(d) The Radiation Protection Consultations and Services Project (RPCSP), which was initiated in 1988 at research institutes, medical centres and industrial companies;
(e) For personal dosimetry, the use of film badges (FB) and thermoluminescence dosimeters (TLD) for estimating external exposure and a whole body counter for estimating internal exposure;
(f) Egyptian NORM regulations, issued as Atomic Energy Authority orders for site categorizations — PET(1) issued in 1999 and updated as PET(2) in 2006.

2. RADIATION MONITORING AT OILFIELDS

The scope of the radiation protection consultations and services covers several oil and gas companies in Egypt. These companies and its oilfields are located at the Gulf of Suez and in the Western Desert. Working areas were classified as follows:

(a)	Normal area:	Dose rate <0.5 μSv/h;
(b)	Supervised area:	Dose rate 0.5–3 μSv/h;
(c)	Controlled area:	Dose rate 3–10 μSv/h
(d)	Restricted area:	Dose rate >10 μSv/h.

By periodic gamma monitoring at various sites (even around pipes entering the process plant), it is possible to distinguish between oilfields that are free from NORM and those that are not. The formation of NORM scale inside pipes (see Fig. 1) leads to a decrease in the production rate. In order to upgrade an old oilfield, several contaminated units have to be replaced by new units. In the present study, attention was paid to personal dosimetry issues.

3. DOSIMETRIC RESULTS

Petroleum companies became involved in the cleaning of NORM contaminated units through contracts with several Egyptian service providers. Workers were medically examined and provided with film badge dosimeters. All necessary health, safety and environmental measures, including measures for radiation protection, were taken when handling NORM contaminated units. In the present study external gamma doses were assessed for (1) workers completing their work within two months (early phase), and (2) workers carrying out their work in up to seven periods.

FIG.1. NORM contaminated pipe.

3.1. Case 1: Workers who completed their work within two months

The distribution of effective doses from external gamma exposure is given in Table 1. A breakdown of the effective doses received by workers from different contracting companies is given in Table 2.

TABLE 1. WORKER DOSE DISTRIBUTION

Effective dose interval (mSv)	Number of workers	Proportion of total (%)
0–0.5	148	77
0.5–1.0	24	12
1.0–1.5	11	6
1.5–2.0	3	2
2.5–3.0	5	3

TABLE 2. EFFECTIVE DOSE RECEIVED BY WORKERS, BY CONTRACTING COMPANY

Contractor number	Number of workers	Effective dose (mSv)			Collective effective dose (man·mSv)
		Minimum	Maximum	Average	
1	82	0.1	1.7	0.3	24.8
2	24	0.2	2.2	0.3	7.2
3	20	0.1	2.1	0.45	9.0
4	16	0.1	2.3	0.45	7.2
5	15	0.1	0.3	0.2	3.0
6	15	0.1	1.1	0.38	5.7
7	6	0.1	0.6	0.27	1.6
8	4	0.1	0.3	0.225	0.9
9	3	0.1	0.2	0.2	0.6
10	2	0.05	0.1	0.1	0.2
11	2	0.3	1.1	0.7	1.4
12	1	—	—	0.3	0.3
13	1	—	—	0.2	0.2
14	1	—	—	0.2	0.4
Total	192	0	2.3	0.35	65.9

3.2. Case 2: Workers who carried out their work over several periods

The doses from external gamma exposure received by workers in each period are given in Table 3. The dose distribution for a period of five months is given in Table 4. The doses for different periods are given in Table 5.

4. CONCLUSIONS

From the results of the present work, the average external gamma dose received by a worker was 0.6 mSv over a five month period. Nearly 80% of the workers received gamma doses of less than 1 mSv, while the remaining 20% received doses greater than 1 mSv (up to 3 mSv).

TABLE 3. EFFECTIVE DOSE RECEIVED BY WORKERS COMPLETING UP TO SEVEN WORKING PERIODS

Period	Number of workers	Effective dose (mSv)	
		Average	Range
1	29	0.15	0.05–1.0
2	34	0.14	0.05–1.0
3	26	0.15	0.05–1.4
4	37	0.15	0.05–0.7
5	30	0.05	0.05–0.3
6	30	0.23	0.05–0.9
7	6	0.22	0.05–0.5

TABLE 4. WORKER DOSE DISTRIBUTION OVER A FIVE MONTH PERIOD

Effective dose interval (mSv)	Number of workers	Proportion of total (%)
0 to 0.5 mSv	463	59.36
0.5 to 1.0 mSv	155	19.87
Sub-total (less than 1 mSv)	618	79.24
1.0 to 1.5 mSv	100	12.82
1.5 to 2.0 mSv	38	4.87
2.0 to 2.5 mSv	15	1.92
2.5 to 3.0 mSv	9	1.15
Sub-total (greater than 1 mSv)	162	20.76
Total	780	100

TABLE 5. WORKER DOSES FOR DIFFERENT PERIODS

Period (d)	Number of workers	Collective dose (man·mSv)	Average individual dose (mSv)
32	236	118	0.5
113	748	271	0.37
124	767	382	0.49
154	780	482	0.62

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OCCUPATIONAL EXPOSURE ASSESSMENT AT A TITANIUM DIOXIDE PLANT LOCATED IN THE SOUTH-WEST OF SPAIN

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Abstract

Within the framework of a broad project devoted to the control of exposures in several typical NORM industries located in the south of Spain, occupational radiation exposures in one industry devoted to the production of titanium dioxide pigments using the sulphate process have been evaluated. External (gamma) and internal (inhalation) pathways of exposure were considered in the evaluation, arriving at the conclusion that, under normal operating conditions, the incremental dose due to occupational exposures is less than 1 mSv/a. However, certain precautions should be taken during some maintenance operations, while any drastic changes in the worker occupancy periods within the various areas of the factory should be avoided without a prior radiological evaluation.

1. INTRODUCTION

A facility for the production of titanium dioxide pigments is one of several chemical industries located near Huelva in south-western Spain. The facility processes large amounts of ilmenite containing elevated concentrations of radionuclides of natural origin and can therefore be regarded as a typical NORM industry in which it is necessary to evaluate the possible incremental exposures of workers and the public [1]. The results of the evaluation will determine whether this industrial activity requires radiological surveillance and, if necessary, corrective actions to reduce the exposures and/or the application of radiation protection measures.

The main conclusions from the assessment of occupational exposure at the Huelva titanium dioxide industry are presented. In considering the characteristics of the industrial process, external gamma radiation and dust inhalation were identified as the important exposure pathways. Other exposure pathways such as ingestion or material deposition on the skin were considered to be insignificant.

Representative process materials such as feedstock, intermediate materials, residues and co-products were sampled and their activity concentrations determined by alpha spectrometry (^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{210}Po) or gamma spectrometry (^{226}Ra, ^{228}Ra, ^{228}Th). Essential knowledge about the behaviour of the various radionuclides throughout the process was gained from these determinations, providing important baseline information for the mapping of external gamma dose rates within the factory.

Spot measurements of external dose rate were made at various locations within the plant, using a Berthold LB1236 survey meter sensitive to photons in the 0.02–1.2 MeV energy range. These measurements were compared with values obtained using the same equipment outside the facility, in areas where dose rates were considered to be at background level. In this way, it was possible to evaluate the incremental gamma dose rates arising from the industrial process.

For the estimation of the inhalation doses, air sampling filters were deployed in the dustiest zone of the factory using a high volume sampler (40 m^3/h). Additionally, and for reference purposes, air sampling filters with the same experimental system were deployed in an area not affected by industrial activities, located 30 km from the titanium dioxide plant. The activity concentrations of ^{210}Pb on the filters were determined by gamma spectrometry. The activity concentrations of the alpha emitters (U isotopes, Th isotopes, ^{226}Ra and ^{210}Po) were determined by alpha spectrometry.

2. TITANIUM DIOXIDE PIGMENT PRODUCTION PROCESS

Titanium dioxide is a simple inorganic compound produced as a pure white powder for use as a pigment in products such as paints, plastics, high grade papers and printing inks. The titanium dioxide product is not radioactive, but the feedstock contains elevated concentrations of radionuclides of natural origin. Pure titanium dioxide is produced by either the ‘chloride’ or ‘sulphate’ process route. The sulphate process route (with ilmenite mineral as the feedstock) is used at the Huelva facility and is illustrated in Fig. 1.

The titanium dioxide plant imports ilmenite rock as a feedstock, composed mainly of TiO_2, FeO and Fe_2O_3, and in a first step is dried and milled. The milled raw material is then digested with 98% H_2SO_4 supplied by other local industries together with 80% H_2SO_4 recycled from later stages of the process. As a result of

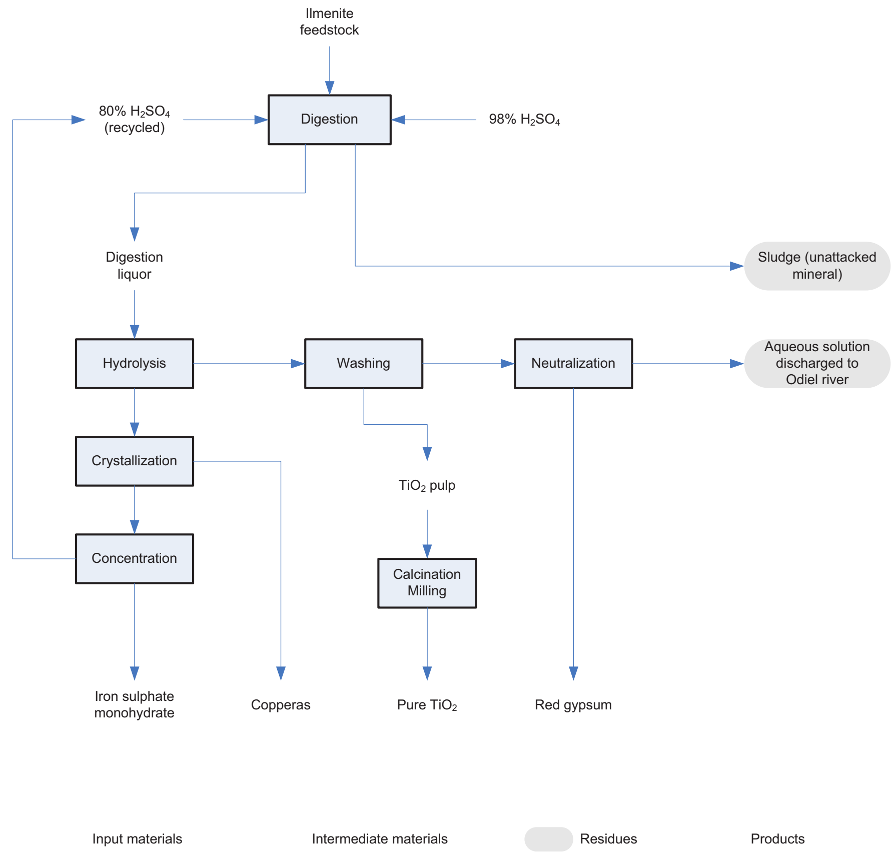

FIG 1. Sulphate process route.

the digestion, a liquor containing the majority of the Ti and Fe in solution is formed, while a sludge containing undigested material is separated from the process as a waste.

The liquid from the digestion is then hydrolysed, causing the precipitation of a TiO_2 pulp containing some impurities and the generation of a strong acid liquid fraction enriched in Fe. After precipitation, the hydrated TiO_2 is separated from the strong liquid acid by vacuum filtration. The strong acid liquid fraction is subjected to an initial crystallization process where crystals of ferrous sulphate heptahydrate (copperas) are formed as a co-product. This is followed by concentration and maturation of the resulting supernatant, resulting in the precipitation of another co-product, iron sulphate monohydrate. The liquid remaining after these steps is H_2SO_4, which is evaporated until it reaches the strength needed for recycling into the digestion process.

The TiO_2 pulp separated after the hydrolysis step is washed and filtered several times. The final solid cake is then subjected to calcination and other physical processes in order to produce TiO_2 pigment in different sizes and qualities. These physical processes are not described because they do not have any radiological implications (the washed solid cake is practically free of radionuclides, as shown later in the paper).

Finally, the slightly acid solution formed as a result of the washing of the TiO_2 pulp is neutralized, resulting in the precipitation of a red gypsum co-product containing the majority of the residual metals and radionuclides. The neutralized liquid fraction obtained after separation by filtration of the red gypsum is released to the nearby Odiel River and thence to the sea.

In summary, therefore, in addition to the commercial final product (TiO_2 pigment), three co-products (copperas, iron sulphate monohydrate and red gypsum) and one residue (sludge containing undigested material) are generated, while neutralized liquid is released to the sea.

3. RADIONUCLIDE FLUXES

The results of radionuclide activity concentration measurements for the feedstock, intermediate materials, co-products, waste and releases associated with the plant are given in Table 1. From these results, and knowing the annual

TABLE 1. ACTIVITY CONCENTRATIONS IN SAMPLES OF PROCESS MATERIAL

	Activity concentration (Bq/g)				
	^{238}U	^{226}Ra	^{232}Th	^{228}Ra	^{228}Th
Ilmenite feedstock	0.088±0.004	0.098±0.005	0.359±0.020	0.303±0.018	0.322±0.011
Sludge	0.312±0.019	0.877±0.021	0.373±0.012	2.616±0.069	0.704±0.017
Copperas	0.0009±0.0001	ND[a]	0.008±0.0002	ND	0.0091±0.0004
Iron sulphate monohydrate	0.053±0.001	0.011±0.001	0.365±0.006	0.044±0.002	0.411±0.012
Red gypsum	0.0194±0.0004	0.014±0.001	0.115±0.002	0.088±0.003	0.122±0.003
Effluent	<0.0001	<0.0001	<0.0001	<0.000l	<0.0001
Digestion liquor	0.018±0.001	0.003±0.001	0.130±0.008	0.020±0.002	0.120±0.009
TiO_2 pulp	0.003±0.001	0.006±0.001	0.005±0.001	0.025±0.001	0.008±0.001

[a] ND = not detectable

consumption and production of the raw materials, co-products and waste, the following conclusions can be drawn:

(a) In the feedstock mineral there is secular equilibrium between ^{238}U and its progeny and between ^{232}Th and its progeny. Within the process, the ^{232}Th series radionuclides account for three to four times more activity than the ^{238}U series radionuclides.

(b) The sludge is particularly enriched in radium isotopes, containing about 90% of the total radium introduced to the process. This sludge also contains a significant proportion (30%) of the uranium introduced to the process and a minor proportion of the thorium isotopes.

(c) Most of the thorium and a high proportion of the uranium report to the liquor formed in the digestion phase and eventually end up mostly in the iron sulphate monohydrate co-product. On the other hand, the copperas co-product is practically free of radioactivity.

(d) After liberation from the pulp in the washing steps, some residual (but nevertheless significant) amounts of thorium and uranium isotopes end up in the red gypsum co-product. The neutralization process is very effective in removing radionuclides from the liquid washing fraction, such that the effluent released to the sea is essentially free of radioactivity.

(e) The TiO_2 pulp contains low concentrations of radionuclides with no significant radiological impact.

The conclusions drawn from these results are in agreement with those found in other, rather scarce, literature on the subject [2] and are in accordance with the results of separate exposure studies conducted in the plant, which are summarized in the following sections.

4. EXPOSURES ASSOCIATED WITH THE MANAGEMENT OF FEEDSTOCK, CO-PRODUCTS AND RESIDUES

An excellent correlation was found between the radiometric determinations performed on the feedstock, co-products and waste and the external gamma dose rates measured in the areas of the factory where these materials are stored. The external gamma dose rates determined from spot measurements in these storage areas are given in Table 2.

The dose rates measured in storage areas for copperas and red gypsum are essentially at background levels, in line with the low content of gamma emitters in these co-products. Higher values (up to eight times background levels) were found in the sludge storage area due to the presence of high concentrations of radium isotopes and their progeny in the sludge. Gamma dose rates in the iron sulphate monohydrate storage area were low, despite the relatively high overall radioactive content of this co-product, because the material contains low concentrations of radium isotopes, which are the principal gamma emitters.

It should be noted that the generation of certain co-products within the process is a consequence of reducing the impact of possible releases of metals and radionuclides to the environment. The production of iron sulphate monohydrate from the effluent leaving the crystallization section allows the recycling of the remaining acid solution and avoids the need for treating these effluents as residues (and thus avoiding any possible public impact). The red gypsum is generated for the removal of the acidity in the water used for washing of the TiO_2 pulp before its release to the sea. This results in a drastic reduction in the concentration of heavy metals and radionuclides in the effluent. The company has found applications and markets for most of the generated co-products. The copperas is used, for example, for the recovery of basic soils in agriculture, as a component in animal feeding or as a flocculant in water treatment, while the iron sulphate monohydrate is used as a raw material in the production of ferric fertilizers for the treatment of iron-deficient agricultural soils. The only material without a defined market is the sludge produced in the digestion of the feedstock. This is transported to a residue management plant where it is converted into an inert residue and stored.

As a result of the management strategy of the company, the direct environmental and public radiological impact is essentially zero, while the radiological impact associated with the distribution and use of the co-products is minimal because of the very low radioactive content (see Tables 1 and 2). In the case of workers involved in the management of the sludges inside the factory, the incremental gamma dose rate is about 0.5 μSv/h (see Table 2) and the occupancy period is 200 h/a. This gives an incremental effective dose due to external gamma

TABLE 2. GAMMA DOSE RATES AT STORAGE AREAS

	Dose rate (μSv/h)
Open area for storage of ilmenite (7 measurements)	0.22–0.39
Open area for storage of sludges (6 measurements)	0.56–0.63
Storage area for copperas (3 measurements)	0.08
Storage area for iron sulphate monohydrate	0.14
Storage area for red gypsum	0.12
Outside the factory (background)	0.08–0.09

exposure of 0.1 mSv/a. The possible incremental dose received through inhalation can be considered negligible, because these sludges are generated, stored inside the factory and finally transported to the treatment plant in wet form.

5. OCCUPATIONAL EXPOSURES IN DIFFERENT AREAS OF THE PLANT

The gamma dose rates measured in various areas of the plant are generally quite moderate, as shown in Table 3, especially when taking into account the background dose rate, which was determined to be about 0.1 μSv/h in a non-contaminated area outside the factory. Considering the radiometric results shown in Section 3, the explanation is simple because only U, Th and other isotopes that contribute minimally to the gamma dose rate remain with the process flow, while most of the radium leaves the process flow with the undigested material immediately after the digestion of the feedstock.

Limited instances of higher dose rates were found in certain areas. These can be attributed to the presence of residual amounts of radium isotopes that, while following the process flow in quite low concentrations, are precipitated and deposited onto the internal surfaces of tanks and pipes, where they accumulate as scale. This fact could seem surprising given that the majority of the radium isotopes leave the process at the sludge formation stage of the digestion process, but it is their marginal solubility that causes them to precipitate and deposit while following the process flow as a result of variations in physical conditions such as pressure. Scale formation occurs particularly where the liquor resulting from the precipitation of the TiO_2 is crystallized for the formation of the copperas — an abrupt decrease in pressure provokes the ebullition of the liquor at low temperature and the consequent formation of the copperas crystals. Concentrations of ^{226}Ra and ^{228}Ra of the order of 100 Bq/g have been measured in scales collected from the crystallizers. Elevated gamma dose rates have also been measured in the hydrolysis area, and are associated with the formation of scales and with the fixation of certain amounts of radium isotopes in the filter clothes used for the separation of the TiO_2 pulp from the strong acid liquor. These dose rates increase with the time of use of the filter cloths.

The elevated gamma dose rates given in Table 3 do not imply that workers necessarily receive effective doses that are high enough to warrant specific radiation protection measures under normal operating conditions. In the majority of the areas of the plant, the dose rates are quite moderate, while the higher values are obtained in limited zones that either are not accessible to the workers or are characterized by a very low occupancy factor. For example, it has been estimated that under normal operating conditions a worker spends less than 50 h/a in the

TABLE 3. GAMMA DOSE RATES AT VARIOUS AREAS WITHIN THE PLANT

	Dose rate (μSv/h)
Open area for storage of ilmenite	0.22–0.39
Milling area	0.12–0.25
Digestion area	0.11–0.23
Sludge separation area	0.10–0.23
Hydrolysis area	0.10–0.66
Crystallization area	0.15–12.60
Maturation area (iron sulphate monohydrate)	0.10–0.55
Washing area	0.15–0.28
Neutralization area (red gypsum)	0.10–0.15
TiO_2 pigment finishing area	0.10–0.12

vicinity of the crystallizers where the maximum dose rates were observed, implying an effective dose of less than 0.5 mSv/a. However, precautions should be taken in these limited areas during maintenance operations, particularly if the scales need to be removed to avoid interference with the production process.

Finally, it should be noted that the committed effective dose received by a full time (2000 h/a) worker due to the inhalation of airborne dust is much lower than the effective dose arising from external gamma exposure. An investigation revealed that resuspension of dust gave rise to an average airborne dust level of 140 ± 50 $\mu g/m^3$, significantly higher than the value of 19 ± 7 $\mu g/m^3$ measured at the reference background station. The resuspended material has similar Th series and U series radionuclide concentrations as the ilmenite feedstock; this is to be expected, given that the storage and milling of ilmenite is the main process by which dust becomes resuspended. On this basis, the average incremental dust activity concentration in air would be less than 50 $\mu Bq/m^3$. This implies an incremental committed effective dose by inhalation of the order of 10 μSv/a.

6. CONCLUSIONS

The assessment of occupational exposures in a titanium dioxide pigment plant located in south-western Spain has been reported and discussed in this paper. Under normal operating conditions, the incremental effective doses received by workers have been estimated to be less than 1 mSv/a, with the major contribution coming from external gamma exposure. The management policy of the company regarding co-products and waste has also been highlighted, in that it has brought about a drastic reduction in the environmental impact and the radiological impact on the public without increasing the radiological impact on the workers. The company has found applications and markets for most of the co-products generated, decreasing at the same time the impact of their releases to the environment.

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OPERATIONAL RADIATION PROTECTION ASSOCIATED WITH THORIUM PROCESSING IN INDIA

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Abstract

Thorium concentrate is produced from the mineral monazite obtained from beach sand mining and mineral separation. The radiological issues associated with the production of thorium in different chemical forms, purification and related waste generation and disposal in a typical Indian plant are discussed. The chemical processes involve operational radiological safety problems of varying magnitudes. The generation of the different solid wastes, liquid effluents and gaseous releases are identified and quantified. The methods of treatment, conditioning and disposal of these wastes are indicated. Radiological properties and hazards of the radionuclide and emissions involved are identified with respect to each process step. The gamma dose rates, activity concentrations in air and contamination levels encountered in the work environment at different process stages and problems unique to handling of large quantities of NORM are discussed. The engineered safety features of the process plants and their effect on individual exposures are analysed. The paper also gives a history of the individual exposures resulting from nearly 50 years of operational monitoring, analyses the trend and quantifies the individual exposures by normalizing them with respect to unit product output. The per capita yearly average exposures were in the range of 3.0–7.8 mSv during the processing of monazite. Representative exposures for rare earths production were in the range of 0.3–1.0 man·mSv (mean 0.6 man·mSv) per tonne of rare earth concentrate produced. In terms of thorium hydroxide production, the dose was approximately 6.2 man·mSv per tonne of thorium hydroxide (100%) produced from monazite. For wet thorium hydroxide concentrate sludge the representative exposure works out to approximately 2–2.5 mSv per tonne. Process modifications, decommissioning of the old rare earths plant and commissioning of a new rare earths plant resulted in a reduction in doses during the 1980s, whereas the average doses steadily increased during the 1990s due to additional process changes introduced. Internal exposures accounted for 50–70% of the total dose. Contamination of plant and equipment, a reduction in available ventilation due to the introduction of newer process equipment, and proximity of the thorium storage silos have resulted in higher activity concentrations in air and hence internal exposures. Recently, during the retrieval and processing of thorium hydroxide

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concentrate for the production of thorium oxalate and recovery of uranium, the individual annual exposures were in the range of 5.4–6.2 mSv and the representative exposure for thorium oxalate production works out to be 0.9 man·mSv per tonne of thorium oxalate produced.

1. INTRODUCTION

Monazite, the main mineral of thorium, is found along with the other heavy minerals such as ilmenite, zircon, rutile, garnet, sillimenite in the beach sands of western and eastern coastal India and some inland placers. The mining and mineral separation of these sands and chemical processing of monazite for the production of thorium and rare earth compounds involve occupational and environmental radiological safety problems of varying magnitudes. This paper highlights the operational radiation protection associated with the chemical processing of monazite and thorium based on the vast experience gained over the years of operation of the plants. The plants have been in operation for the last half a century for the production of rare earths and thorium and later on for the recovery of uranium from monazite. The scenario involves large scale handling of thorium concentrates and the related storage and disposal of wastes issues, and the resultant occupational as well as environmental exposures.

2. THE PROCESS

2.1. Chemical processing of monazite

Monazite, an orthophosphate of thorium and rare earth (RE) elements, is chemically processed by reaction with caustic soda lye. The reacted mass which contains tri-sodium phosphate and hydroxides of thorium, uranium and rare earths along with unreacted monazite is leached with water, when tri-sodium phosphate gets dissolved leaving behind the hydroxides. The tri-sodium phosphate (TSP) by-product is converted into crystals and sold. The mixed hydroxides of thorium, uranium and rare earths are reacted with HCl at a controlled pH, when RE remains in solution as $RECl_3$ and the hydroxides of Th and U and the undissolved and unreacted sand remain in the sludge. This sludge is further dissolved in HCl at a higher pH. The unattacked ‘muck’ is separated by filtration and disposed of as active waste. During the above process the radium (^{228}Ra) and lead present in the monazite appear in the rare earth composite chloride ($RECl_3$) fraction and are removed from the product by deactivation and lead elimination by precipitating barium sulphate and lead sulphide. The solid radioactive waste thus obtained, termed as mixed cake, is disposed of as active waste. The deactivated and lead-free composite rare earths chloride is evaporated and made into flakes and marketed. For the separation of individual rare earths, a solvent extraction process is employed. The diversified products produced include oxides, fluorides and carbonates of composite rare earths, cerium compounds, salts of the heavy rare earths Sm, Gd, Eu and Y and compounds of the light rare earths Nd and Pr. The uranium present in the thorium fraction is recovered by a solvent extraction method.

2.2. Processing of thorium hydroxide concentrate

In earlier periods of operation, thorium hydroxide was stored in silos without separating the undissolved material and uranium contained within it. During recent years, the stock of thorium hydroxide is being retrieved from the storage silos and processed for recovering uranium values and the thorium is converted to thorium oxalate and re-stored in engineered concrete storages. The process adopted is dissolution of thorium hydroxide concentrate in HCl, recovery of uranium by solvent extraction using alamine as a solvent and purification and precipitation of thorium as thorium oxalate. The insoluble muck is filtered out and disposed of as active waste and thorium oxalate is re-stored in engineered concrete silos.

3. OPERATIONAL RADIATION PROTECTION

3.1. External radiation exposures and control

Handling of monazite, thorium concentrates and related process wastes poses external as well as internal radiation hazards. The high energy betas and gammas emitted by the decay products of thorium present in the minerals and chemical concentrates results in elevated external radiation fields in the work environment. The typical external exposures encountered at the different processing stages are given in Table 1. In mining and mineral separation the dose rates largely depend on the monazite content of the feed sand. The dose rates are low except in areas having a presence of monazite or monazite enriched sand fractions. At the plants processing monazite chemically, the dose rates are comparatively higher. Dose rates of the order of 1000 μGy/h are encountered in the deactivation of rare earths chloride concentrates. Freshly separated thorium gives rise to a dose rate of 70–150 μGy/h on contact with bulk material and the dose rates build up over time to values nearly four times higher. The dose rates inside thorium concentrate storage vaults are in the range of 500–700 μGy/h and

TABLE 1. EXTERNAL EXPOSURES ASSOCIATED WITH THORIUM PROCESSING

	Absorbed dose rate (μGy/h)	Ref.
Mining	1–4	[1]
Mineral separation	1–30	[2]
Monazite digestion	0.5–50	[2]
Th, RE, U extraction	2–600	[2, 3]
Ammonium diuranate production	0.3–15	[3, 4]
Thorium oxalate production	25–220	[4]
Deactivation of rare earths	100–1100	[4]
Rare earths compounds production	0.2–120	[4, 5]
Thorium concentrate bags	200–230	[4]
Ammonium diuranate drums	1.5–2.0	[4]
RE concentrate drums	1–2	[4]
TSP bags	<1	[4]
Thorium storage	200–700	[6]
ThO_2 production	30–100	[4]

depend on the age of thorium since chemical separation. Handling of fresh ThO_2 involves dose rates in the range of 100–150 μGy/h.

External exposure control is effected by the judicious application of the basic concept of controlling time, distance and shielding in the design and operation of facilities and processes. The control measures are broadly zoning of work areas based on the contamination potential, segregation of active, less active and inactive process streams, shielding for active material bins, reaction vessels, storage tanks etc., prevention of spillages and prompt removal of spillages from working areas, mechanized handling, special work permits to regulate exposures and job rotation for workers at certain operations.

3.2. Internal exposures and control

In thorium process plants, internal hazards are mainly by way of inhalation of particulates bearing isotopes of thorium and uranium isotopes and daughter products of gaseous thoron (^{220}Rn). Among the progeny nuclides of thoron, ^{212}Pb and ^{212}Bi mainly contribute dose to the lung. Hence the potential alpha energy concentrations due to thoron and progeny are routinely monitored in thorium process plants. The inhaled particulate activity gets deposited in different regions of the respiratory tract, depending on the particle size. A portion of the finer dust, termed as the respirable fraction, gets deposited in the pulmonary region where it stays and irradiates the surrounding tissue. A portion of this dust is solubilized and transported to various body tissues through body fluids. The major site of permanent deposition of thorium is the bone surface where it has a long effective half-life.

Ingestion by way of thorium intake by contaminated food or drinking water is another potential route of exposure. The nuclides get into the body by gastro-intestinal absorption. The absorption is very poor for thorium (10^{-4}) whereas it is 20% for radium. Table 2 gives typical airborne conentrations encountered at different stages in thorium processing. Sources of airborne dust and airborne activity in the facilities are mainly due to spillages, floor and equipment contamination and resuspension, thoron emanation from different matrices, accumulations of active materials, loss of containment, inadequate air changes, ventilation deficiencies, poor housekeeping and accidents.

The reaction vessels are connected to a ventilation system which exhaust through a caustic soda scrubber and chimney. General ventilation in the plant is provided by the wall mounted extractors provided in addition to the ventilation system. The chances of airborne activity concentrations exceeding the derived limits have to be avoided by provision of adequate ventilation. Engineered controls and administrative control measures are adopted for keeping the internal exposures as low as reasonably achievable (ALARA). Some of the measures to control internal exposures include good housekeeping and adoption of best practices,

TABLE 2. AIRBORNE CONCENTRATIONS ASSOCIATED WITH THORIUM PROCESSING

	Airborne concentration			
	^{232}Th (Bq/m^3)	Thoron gas (Bq/m^3)	Thoron progeny (mWL)	Reference
Mineral separation	0.0004–0.11	100–300	0.5–30	[2]
Monazite digestion	0.002–0.22	300–2000	5–300	[2]
Th/RE/U extraction	0.002–0.59	2000–50 000	25–1900	[2]
Thorium oxalate production	0.07–0.59	2000–50 000	30–1800	[4]
Deactivation of rare earths	0.01–0.07	2000–50 000	45–425	[4]
RE compounds production	0.002–0.09	300–1000	5–160	[4]
Thorium storage (silos)	0.01–2.0	10^5–10^6	20 000–100 000	—
ThO_2 production	0.001–0.10	300–2000	100–300	—

avoiding active material accumulations, provision of adequate air changes by way of a good ventilation system comprising of supply–exhaust blowers, ducting, scrubbers or HEPA filters and stack, job rotation for workers in different plants or different locations within plants, mechanization of operations, the use of personal protective equipment and special work permits to control exposures.

3.3. Occupational radiation exposures

At the plant processing monazite, the per capita yearly average exposures were in the range of 3.0–7.8 mSv during the processing of monazite for a half century period until 2002. Process modifications, decommissioning of the old rare earths plant and commissioning of a new plant for processing rare earths have resulted in a reduction of the doses during the 1980s [3]. However, the average doses steadily increased during the 1990s due to new process streams introduced and the generation of an additional quantity of unattacked monazite waste and accumulation of active material. Internal exposures accounted for 50–70% of the total dose. Contamination of plant and equipment, a reduction in available ventilation due to the introduction of newer process equipment, and proximity of the thorium storage silos have resulted in higher airborne activity concentrations and hence internal exposures. The representative exposures for rare earth production were in the range of 0.30–1.00 man·mSv (mean = 0.64, SD = ±0.19) per tonne of rare earth concentrate produced. This dose can also be expressed in terms of other products as well. For wet thorium hydroxide concentrate sludge, the representative exposure works out to approximately 2.0 mSv per tonne. During the retrieval and processing of thorium hydroxide concentrate for the recovery of uranium and production of thorium oxalate, the individual annual exposures were in the range of 5.4–6.2 mSv and the representative exposure for thorium oxalate production worked out to 0.90 man·mSv per tonne of thorium oxalate produced.

During the transport of thorium concentrate to different destinations, the per capita radiation exposure received by the truck crew members ranged from <0.2 to 4.4 mSv per trip, depending on the destination. The material is transported as LSA material duly labeled as full load (exclusive use) in trucks with relevant documents. The crews are given instructions in writing to meet any emergency and an emergency kit is sent along with each consignment.

3.4. Waste management and environmental surveillance

The chemical processing of the minerals produces solid wastes of moderately higher activity concentrations and substantial quantities, requiring careful planning and execution of the waste disposal operations. The details of the wastes generated in the plant are given elsewhere [1]. There are three kinds of solid wastes, namely insolubles and unreacted sludge, mixed cake, and effluent treatment cake. The significant radionuclide in the waste is ^{228}Ra and its activity concentration is 100–10 000 Bq/g in different wastes. These are disposed of into reinforced cement concrete trenches. The dose rate on contact with the waste is 60–500 μGy/h and the dose rate over the sealed trenches is 2–5 μGy/h.

Thorium concentrates are stored in RCC Silos at the plant site. These are engineered structures with adequate shielding which ensures that there is no enhancement of the external radiation background in public areas. The thoron and its progeny nuclides decay inside the silo itself. Release of airborne activity takes place mainly from processing plants and the effluents are discharged through stacks. The reaction tanks and storage tanks are connected to the main ventilation system, which also incorporates a caustic soda scrubber. The estimated annual releases are 70–360 GBq for thoron progeny. The liquid effluents are properly treated to contain the radionuclides, especially ^{228}Ra, prior to discharge into the river. The annual discharged activity is of the order of 0.5–1 GBq.

Regular radiation surveys are carried out in the plant premises and public areas surrounding the plant. Area monitoring TLDs are posted in the environment to estimate the exposures. The radiation exposures in some of the areas adjacent to the factory compound wall range from 0.40 to 1.80 mSv per year, and corresponding natural background radiation exposures estimated up to a distance of 35 km from the site ranged from 0.2 to 1.4 mSv per year [2, 7]. Monitoring for radioactivity in environmental air samples has been undertaken on a regular basis. Analysis indicated that there is no significant impact of these nuclides beyond 300 m from the plant boundary. The per capita committed effective dose due to the inhalation of Th and progeny works out to about 10–30 μSv per year for the representative persons in the vicinity. Monitoring of the aquatic environment, including groundwater, around the waste disposal trenches did not reveal any enhancement in activity concentration over the years.

4. CONCLUSIONS

The chemical processing of monazite and thorium involves operational radiological safety problems of varying magnitudes. These parts of the front end of the nuclear fuel cycle involve average per capita occupational exposures in the range of 3.0–7.8 mSv per year. Representative exposures for rare earth production were in the range of 0.3–1.0 man·mSv with a mean of 0.6 man·mSv per tonne of rare earth concentrate produced. In terms of thorium hydroxide production, the dose was approximately 6.2 man·mSv per tonne of thorium hydroxide (100%) produced from monazite. For wet thorium hydroxide concentrate sludge, the representative exposure works out to approximately 2–2.5 mSv per tonne. Recently, during the retrieval and processing of thorium hydroxide concentrate for the production of thorium oxalate and recovery of uranium, the individual annual exposures were in the range of 5.4–6.2 mSv and the representative exposure for thorium oxalate production worked out to be 0.9 man·mSv per tonne of thorium oxalate produced. Some 40–50% of the exposure accounts for internal dose. Environmental exposures of the public resulting from the chemical processing of monazite are not significant. However, regular monitoring is required to ensure protection.

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RAPPORTEUR SUMMARY OF OPENING SESSION AND TOPICAL SESSION 1

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1. INTRODUCTION

This report briefly reviews the papers delivered in these sessions, while concentrating mainly on the topics that appear to have greater importance for the NORM community and on common themes from the presentations.

2. OPENING OF THE SYMPOSIUM

The addresses given at the opening of the symposium reminded delegates of the need to protect people and the environment from the potentially harmful effects of exposure to NORM. The symposium has come to Morocco at a time when this country's interest in nuclear issues is increasing. Morocco has a 2 MW research reactor and is also currently hosting a conference in Marrakesh on the Safety of Research Reactors. Morocco has the largest phosphate deposits in the world. The exploitation of these deposits involves exposure to NORM, making Morocco a most appropriate location for NORM VI.

3. DEVELOPMENTS AT THE INTERNATIONAL LEVEL

The revision process for the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Sources (the BSS) was the subject of the Keynote Address. Drafting of the revised BSS was nearing completion after more than three years of work. The presentation outlined the structure of the publication, the review process and the involvement of the various sponsoring organizations. New or changed requirements were being introduced in several areas, including exemption and clearance, non-medical imaging and radiation generators and sources. The categorization of exposure situations into 'planned exposure situations' and 'existing exposure situations', and the implications of this for exposure to natural sources, was discussed in some detail. As with the current BSS, most exposures to natural sources were not treated as practices and were therefore, by default, subject to the requirements for existing exposure situations. Exceptions were made, however, for situations having the characteristics of practices, such as industrial activities involving NORM, and these were subject to the requirements for planned exposure situations. In such cases, consideration had to be given to exemption and clearance of material, for which additional numerical criteria were now being introduced.

The current international interest in exposure to radon was also mentioned. The results of recent studies have led the International Commission on Radiological Protection (ICRP) to believe that the cancer risk associated with radon is higher than previously thought. This has caused the ICRP to revise the criteria for radon exposures in homes and workplaces. Similar conclusions have been drawn by the World Health Organization (WHO) in its newly-published Radon Handbook. New dose conversion factors are needed for the full implementation of the new risk estimates into the system of radiation protection, but their availability from the ICRP appears to still be 1–2 years away.

4. DEVELOPMENTS AT THE NATIONAL LEVEL

Progress in implementing the European Council Directive 96/29/Euratom was reported from France. NORM had been investigated extensively in several types of industry. The investigation had included 4200 gamma spectrometry analyses. One finding from the investigation was that some operators do not consider all the important exposure pathways when conducting their own examinations of NORM risks. The investigation gave excellent examples of situations in which both external and internal exposure are significant pathways. Doses received by workers had been estimated, initially using very conservative assumptions. Some activities, such as industrial uses of thorium, had been found to result in high doses that warranted more careful consideration. Some new types of NORM industries had been identified as needing examination, such as the paper manufacturing industry.

5. DEVELOPMENTS IN SPECIFIC NORM INDUSTRIES

5.1. The phosphate industry

The Moroccan phosphate mining group Office Chérifien des Phosphates (OCP) has an extensive programme for the measurement of radiological quantities. The local ores typically contain 0.7–1.0 Bq/g of uranium. In the absence of specific Moroccan regulations for NORM industries, OCP has adopted the methodology for the calculation of doses to workers developed in the United Kingdom by the former National Radiological Protection Board (now part of the Health Protection Agency). The results presented showed good evidence of dose optimization. The use of administrative controls and personal protective equipment were discussed.

Poster presentations from Cuba and Jordan discussed risk assessments in the phosphate industry in those two countries, while a poster from Brazil described the innovative use of lichen as bio-indicator to determine contamination effects from nearby mines. Large quantities of residues were produced from the Brazilian phosphate industry and the lichen bio-indicators were used to assess stack emissions of ^{210}Pb as well as resuspension.

5.2. The oil and gas industry

Mechanisms were described by which pipework in the oil and gas industry becomes contaminated with various radionuclides, including Ra, Pb and U. In addition, the decay of radon causes a buildup of ^{210}Po in black powder residues. The oil and gas industry has generated internal company policies which form the basis for advice to the oil and gas industry. Black powder residues are disposed of using a form of injection into particular porous structures remaining after oil extraction is complete.

A review of regulations related to NORM in the Middle East was presented. The paper described an operational approach for dose and contamination estimation for workers upgrading an old petroleum field.

5.3. The titanium dioxide pigment industry

A comprehensive investigation conducted at a titanium dioxide pigment plant in Spain showed that doses received by workers were well below 1 mSv, thus reinforcing the conclusions drawn from a presentation on the same topic at NORM V. Protection was further optimized by the consideration of occupancy factors in different parts of the plant and administrative controls.

5.4. The tantalum industry

A poster presentation from the Tantalum–Niobium International Study Center described the major sources of tantalum, including tantalite and tin slag containing NORM, and highlighted the denial of shipment issue as it applies to the tantalum industry. This problem is similar to that experienced by the uranium mining sector — while NORM activity concentrations were approximately one thousand times lower than in the uranium industry, they were frequently above the transport exemption level. This is a major problem causing considerable expense to the industry for very little, if any, reduction in risk to the community. One solution suggested was for a special-case transport exemption, a concept that is not provided for in the present IAEA Regulations for the Safe Transport of Radioactive Material.

5.5. Industries involving thorium containing minerals

NORM residues from the mining and beneficiation of rare earth ore at Bayan Obo and Baotou in China were the subject of an invited presentation that drew attention to the recycling of such residues rather than their disposal. The paper emphasized that despite waste being the end point of most residues in NORM industries, this need not be the case as there are many potential end uses for such residues. The use of NORM residues in construction materials was managed by diluting with non-radioactive material such that the activity concentration in the final product was low enough not to be of regulatory concern. Residues from the mining and beneficiation of rare earth ore in China are incorporated into cement, building bricks, filling and other construction materials. Ferrous slag is crushed and recycled for iron and waste rock is used for tailings pond construction on the mine site.

The use of old granaries for the storage of large quantities of monazite sand was described in a paper from the Russian Federation. The monazite had been stockpiled since 1960 as a potential source of thorium for use in nuclear reactors. Over the years, the granaries had clearly decayed to sub-standard conditions. New hangars had been constructed over the older wooden structures to ensure adequate isolation of the material. Studies at the storage site had been conducted in response to concerns about health risks to local people and those initially involved in establishing the stockpile. Local ‘legend’ held that the stockpile had resulted in many cancer deaths. The analysis showed that current doses were very small beyond the inner control area, but some workers had received significant doses during the stockpiling of the monazite. Investigations showed that the number of cancer-related deaths among workers was not significantly higher than that in a normal population. This conclusion was supported by estimates based on the collective dose, which again showed no significant excess cancer incidence.

6. MEASUREMENT TECHNIQUES

The work of the Cadi Ayyad University in Marrakesh was highlighted in a series of poster presentations. These studies were based on track etch counting of radon and thoron in sealed vessels. This simple technology was used to great effect to estimate activities once radon and thoron progeny came into equilibrium with their parents. The estimates of U and Th were dependent on secular equilibrium of the top of the decay chains. Studies included method development, doses to the skin, exposure of cave visitors, exposure in baths and radionuclide transfer to honey. Several posters from Spain outlined radiochemical techniques and measurement studies on NORM in a variety of scenarios. A study of NORM risks from the use of welding rods was a particularly interesting example.

7. SUMMARY

The presentations on this first day of the symposium highlighted a number of important issues related to NORM, including:

(a) The need for more international and national guidance on NORM;
(b) The benefits of using NORM residues as valuable by-products in preference to their disposal as waste;
(c) The need for all significant pathways to be considered when conducting dose assessments in NORM industries;
(d) The limited value of activity concentration determinations without a follow-up dose assessment.

REGULATORY ASPECTS

(Topical Session 2)

Chairpersons

R. WIEGERS
Netherlands

J. INCHAOUH
Morocco

LEGAL ASPECTS OF TENORM REGULATION IN THE UNITED STATES OF AMERICA

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Abstract

The paper addresses some of the convoluted issues related to TENORM regulation in the USA. What regulatory body may regulate TENORM and how it is regulated in practice is the topic of the paper. Understanding how the legal concept of' jurisdiction applies to radioactive materials is essential to making sense out of the current reach and limitations of federal and State regulation of TENORM in the USA, and why regulation of natural sources of radiation is uneven and fragmented. As will hopefully become apparent from the paper, the US system of regulating radioactive materials is both qualitative and quantitative — that is, a radionuclide's elemental identity, activity concentration, as well as its origin and processing history.

1. JURISDICTION OVER TENORM

1.1. The basics of jurisdiction

Jurisdiction is the power, right, or authority of a sovereign to govern or legislate (that is, to make the laws and regulations) as well as the power or right to interpret and apply the law. Jurisdiction is limited by the political boundaries of the sovereign authority. Jurisdiction is also personal (authority over a natural or legal person) or subject matter (authority over a thing, such as a radiation source).

In the US legal system, the plenary sovereign power to make national (federal) law is vested in the Congress.[1] Federal law trumps State law.[2] However, those powers that are not expressly delegated to the federal government by the Constitution are reserved to the States.[3] Thus, a State retains the generalized 'police power' to enact laws to protect the safety, health and welfare of its citizens. State legislatures exercise their police power by making statutes and by

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[1] US Constitution, Article 1, Section 1.

[2] US Constitution, Article VI, Clause 2 'the Supremacy Clause'.

[3] US Constitution, Amendment X.

creating regulatory bodies to which rulemaking authority is delegated. The police power of the State, however, is subject to constraints imposed by the federal and State Constitutions, as well as federal and State statutes.

It is important to note that the Supremacy Clause of the Constitution invalidates State laws that "interfere with, or are contrary to the laws of [C]ongress, made in pursuance of the [C]onstitution".[4] Federal law preempts State law explicitly if the language of the federal statute reveals an express Congressional intent to do so.[5] In the absence of such language, the State's law may still be preempted implicitly: 'Field preemption' occurs where the federal scheme of regulation is so pervasive that Congress must have intended to leave no room for the States to supplement it.[6] 'Conflict preemption' occurs where it is impossible to comply with both the federal and State laws, or the State law stands as an obstacle to the accomplishment of Congress's objectives.[7]

1.2. Federal preemption of State regulation of radioactive materials

The federal Atomic Energy Act of 1954, as amended[8] (hereafter, 'AEA'), among other things, defines classes of radioactive materials that are exclusively within AEA jurisdiction ('source, byproduct and special nuclear materials', or, collectively 'AEA materials') and prescribes procedures for licensing, exempting, and otherwise regulating the use of AEA materials "to assure the common defense and security and to protect the health and safety of the public"[9].

It is well settled that the AEA preempts conflicting State attempts to regulate AEA materials. A good example is a challenge to a State issued landfill permit that prohibited the US Department of Energy (DOE) from placing in the landfill "[s]olid waste that exhibits radioactivity above de minimis levels". In citing Supreme Court precedent in *Pacific Gas & Electric v. State Energy Res. Conservation & Dev. Comm'n*, 461 US 190 (1983) the Sixth Circuit Court of Appeals held:

> "As the Supreme Court unequivocally stated in *Pacific Gas & Electric*, "the federal government has occupied the entire field of nuclear safety concerns, except the limited powers expressly ceded to the states." 461 U.S. at 212.

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[4] *Wisconsin Pub. Intervenor v. Mortier*, 501 US 597, 604 (1991) (quoting *Gibbons v. Ogden*, 22 US (9 Wheat) 1, 211 (1824)).

[5] *Barnett Bank of Marion County v. Nelson*, 116 S. Ct. 1103, 1107-08 (1996).

[6] *Id*. at 1108.

[7] *Id*.

[8] 42 United States Code (USC) 2011, *et seq*.

[9] 42 USC 2012.

> Accordingly, the AEA preempts any state attempt to regulate materials covered by the [Atomic Energy] Act for safety purposes. See id. Here, the challenged permit conditions specifically limit the amount of "radioactivity" and ‘radionuclides’ that DOE may place in its landfill. The sources of such ‘radioactivity’ and ‘radionuclides’ are materials covered by the AEA, i.e. source, special nuclear, and byproduct materials. The [State of Kentucky] seeks to impose these conditions to protect human health and the environment. The permit conditions therefore represent an attempt by the [State] to regulate materials covered by the AEA based on the [State's] safety and health concerns, and are thus preempted.”[10]

The *Pacific Gas & Electric* case involved State attempts to limit construction of a nuclear power plant based on safety concerns over spent fuel storage. The Supreme Court based its AEA preemption determination on Section 274(k) of the AEA[11] which states:

> “Nothing in this section [Section 274] shall be construed to affect the authority of any State or local agency to regulate activities for purposes ***other than*** protection against radiation hazards.”

The Court reasoned that “… Congress, by permitting regulation” for purposes other than protection against radiation hazards,” underscored the distinction drawn in 1954 between the spheres of activity left respectively to the Federal Government and the States”.[12]

Thus, if the radioactive material in question is AEA material and in the particular case of natural radionuclides, if the radioactive material in question is uranium or thorium, then state regulation for the purposes of protection against radiation hazards is preempted by the AEA.

Against this backdrop we can begin to analyse how the federal AEA defines a class of naturally occurring radioactive material that is exclusively within AEA jurisdiction, and how conflicts can arise with respect to regulation of technologically enhanced naturally occurring radioactive material (TENORM).

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[10] *US v. Kentucky Natural Resources & Environmental Protection Cabinet,* 252 F.3d 816 (6th Cir. 2001).

[11] 42 USC § 2021(k) (emphasis added).

[12] 461 US at 210.

1.3. The AEA and TENORM

A good starting point is the AEA definition of ‘source material’. The AEA, in conjunction with regulations promulgated by the US Nuclear Regulatory Commission (NRC), defines a class of NORM over which NRC (and those states that have assumed NRC authority over source material pursuant to AEA Section 274b (‘Agreement States’))[13] retains exclusive regulatory jurisdiction. As defined by the AEA:

> “The term ‘source material’ means (1) uranium, thorium, or any other material which is determined by the Commission pursuant to the provisions of section 61 to be source material; or (2) ores containing one or more of the foregoing materials, in such concentration as the Commission may by regulation determine from time to time.”[14]

Pursuant to the above, NRC promulgated a more precise definition of source material in regulations set forth at Part 40 of Chapter 10 of the Code of Federal Regulations (CFR):

> *“Source Material* means: (1) Uranium or thorium, or any combination thereof*, in any physical or chemical form* or (2) *ores which contain by weight one twentieth of one per cent (0.05%) or more* of: (i) Uranium, (ii) thorium or (iii) any combination thereof. Source material does not include special nuclear material.”[15]

NRC commented on the history of the source material definition:

> “The original definition of source material in Part 40 (1947) excluded all forms of uranium and thorium below the concentration limit of 0.05 per cent by weight. In 1961, the definition in Part 40 was revised to its present form, to be consistent with the definition of source material in the AEA of

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[13] AEA Sect. 274b provides that NRC may partially relinquish its authority to a state, upon the state’s meeting certain requirements and formally entering into an agreement with NRC. One necessary requirement is that the Agreement State promulgates regulations that are compatible with those of NRC, including the definition of source material and provisions for exempting unimportant quantities of source material from licensing. Agreement State regulations defining source material and unimportant quantities are virtually verbatim copies of NRC regulations. See, e.g. Georgia Rule 391-3-17-.02, ‘Licensing of Source Material.’

[14] 42 USC 2014(z).

[15] 10 CFR 40.4 (emphasis supplied).

> 1954. Since that time, only ore below the 0.05 per cent by weight concentration has been excluded from the definition of source material. Other forms, 'chemical mixtures, compounds, solutions, or alloys,' in which the uranium or thorium is by weight less than 0.05 per cent of the mixture, became an 'unimportant quantity,' covered by the exemption in § 40.13(a), which was also added to Part 40 in 1961. The concentration appears to have been chosen on the basis of the concentrations of source material necessary to be a useful source of fissionable material."[16]

Importantly, the regulatory threshold of 0.05% is purely a technology based number and is not based on any potential dose from these materials. The absence of any human health considerations in exempting unimportant quantity source material from regulation is an issue that comes into play in the context of NRC jurisdiction over TENORM, discussed later.

Section 62 of the AEA imposes a licensing requirement on persons who transfer or receive source material, along with an express prohibition on licensing *unimportant quantities* of source material:

> "Unless authorized by a general or specific licence issued by the [Nuclear Regulatory] Commission, which the Commission is authorized to issue, no person may transfer or receive in interstate commerce, transfer, deliver, receive possession of or title to, or import into or export from the USA any source material after removal from its place of deposit in nature, ***except that licences shall not be required for quantities of source material which, in the opinion of the Commission, are unimportant."***[17]

Unimportant quantities of source material are defined at 10 CFR 40.13(a) and (b):

> "(a) Any person is exempt from the regulations in this part and from the requirements for a licence set forth in section 62 of the Act to the extent that such person receives, possesses, uses, transfers or delivers source material in any chemical mixture, compound, solution, or alloy in which the source material is by weight less than one-twentieth of 1 per cent (0.05 per cent) of

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[16] SECY-99-259. 'SECY' papers are papers the NRC staff submits to the Commission to inform them about policy, rulemaking, and adjudicatory matters. See www.nrc.gov for SECY papers.

[17] 42 USC 2092 (emphasis supplied).

> the mixture, compound, solution or alloy. The exemption contained in this paragraph does not include byproduct material as defined in this part.
>
> (b) Any person is exempt from the regulations in this part and from the requirements for a licence set forth in section 62 of the act to the extent that such person receives, possesses, uses, or transfers unrefined and unprocessed ore containing source material; provided, that, except as authorized in a specific licence, such person shall not refine or process such ore."

10 CFR 40.13(c) enumerates certain articles and materials containing source material that have been deemed unimportant by NRC. From the perspective of TENORM regulation, the following exempt materials once came into play in the titanium mineral processing context, and could yet arise in the TENORM context:

> "Any person is exempt from the regulation in this part and from the requirements for a licence set forth in section 62 of the Act to the extent that such person receives, possesses, uses, or transfers:
>
> (1) (vi) rare earth metals and compounds, mixtures, and products containing not more than 0.25 per cent by weight thorium, uranium, or any combination of these…"

A petitioner sought to apply the above exemption to wastes generated from titanium ore processing. NRC evaluated the applicability of the above exemption and that of 40.13(a), and concluded the rare earths exemption of 40.13(c)(1)(vi) only applies to rare earth *products*.

> "NRC examined the question of exemption and licensing status for titanium bearing ores and waste products resulting from titanium dioxide manufacturing at a plant in Tennessee. Some ores (monazite and xenotime-rare earth ores) and some waste products (barium salts in scale in piping, and some process wastewater) contain thorium and uranium in excess of 0.05% by weight, but less than 0.25% by weight. It was suggested that these materials were covered by 10 CFR 40.13 (c) (1) (vi) and should, therefore, be exempt from licensing. 10 CFR 40.13 (c) (1) (vi) provides an exemption for licensing for thorium contained in rare earth metals and compounds, mixtures, and products containing not more than 0.25% by weight of thorium, uranium, or any combination of thorium and uranium. This exemption was promulgated in 1961 upon the petition of American Potash and Chemical Company to restore a status quo ante. American Potash was

> then processing rare earth ores for thorium and rare earths at its facility in West Chicago, Illinois. The exemption of 10 CFR 40.13 (c) (1) (vi) can be traced to Schedule I of 10 CFR 40.60. Schedule I was first promulgated in 1947 (12 FR 1855, March 20, 1947) in conjunction with a provision requiring unlicensed persons in possession of 10 pounds of source material ore, or 1 pound of refined source material, to register with the Atomic Energy Commission. However, products listed in Schedule I were exempted. This history indicates that the exemption applies only to products, not to raw materials or process wastes. Further, the petitioner, American Potash and Chemical Company, always proceeded under licence with respect to ores exceeding 0.05% by weight thorium. NRC emphasizes the fact that only products are involved in the several exemptions in paragraph 40.13 (c). Under the regulatory system of 10 CFR Part 40, unrefined and unprocessed ores are exempt without limit on quantity and quality pursuant to paragraph 40.13 (b). If source material ore has been refined or processed . . . it is subject to licensing. 10 CFR 40.13 (c) (9) states that paragraph 40.13 (c) does not authorize manufacturing of any of the products listed in paragraph (c), reinforcing the historical view of the limited application of the exemption to products only, and not to raw materials and waste."[18]

This example shows that the plain language of a regulation, or in this case an exemption from regulation, sometimes must be looked at in the historical context of how the regulation (or exemption) was developed to determine the types of materials to which it applies.

In the titanium ore processing example, it was determined that titanium ores and wastes above 0.05% uranium and thorium are *source material* subject to NRC licensing, notwithstanding their rare earth content. It is important to note that the 'barium salts in scale in piping' very likely contained radium (Ra-226 and Ra-228) in addition to uranium and thorium, because barium and radium exhibit similar chemical properties. This example illustrates a very common occurrence that arises time and again in TENORM regulation: chemical processing of minerals containing natural uranium and thorium can result in *partitioning* of radium to one chemical phase or another. Because virtually *all* materials of natural origin (including petroleum, groundwater, coal, ferrous and non-ferrous ores, etc.) contain some level of natural uranium, thorium, and decay progeny, it is entirely plausible that extractive processes designed to purify and concentrate can, and often do, partition naturally occurring radionuclides to different

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[18] NRC Health Physics Position Paper (HPPOS) 202 (1984).

chemical phases. As will be seen, this partitioning of radium is the basis for one definition of TENORM that has been adopted by some states.

If the titanium processing operation were *licensed* because ores exceeded 0.05% uranium and thorium, then all aspects of ore processing and waste disposal (including radium scales arising from source material processing) would be governed by the terms of the licence and the regulations of the regulatory body that issued the licence (NRC or Agreement State).

On the other hand, what if the titanium ores were ‘unimportant quantities’ of source material below 0.05% uranium and thorium? What if the ‘barium scales’ became substantially enriched in radium content, but were also below 0.05% uranium and thorium? NRC may have AEA *jurisdiction* over unimportant quantities, but unimportant quantities are excluded from licensing by AEA Sect. 62 and 10 CFR 40.13(a). Processing ‘unimportant quantities’ and the unintentionally concentrated radium, therefore is excluded from any form of NRC licensing applicable to AEA materials.[19] So, if regulatory control is to be imposed on the ‘radium scales,’ it is up to a regulatory body other than NRC, one who can lawfully assert jurisdiction over this type of radioactive material: it might be the US Environmental Protection Agency (EPA) if the radium scales come within EPA’s statutory grant of jurisdiction, or it might be the relevant state radiation protection authority, who could assert jurisdiction under the ‘police power’ of a state to enact laws to protect the safety, health and welfare of its citizens.

1.4. Source material versus TENORM

According to the federal AEA definition and NRC’s (or compatible Agreement State’s) cited above, naturally occurring uranium and/or thorium is ‘source material’, but what kind of source material is it?

(a) If the material in question contains less than 0.05% by weight uranium and/or thorium, in any physical or chemical form, including any chemical mixture, compound, solution, or alloy, then it is an unimportant quantity of source material. Section 62 of the AEA prohibits the NRC, and preempts

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[19] The Energy Policy Act of 2005 amended Section 11 e.(3) of the Atomic Energy Act of 1954to include as byproduct material: (i) any discrete source of radium-226 that is produced, extracted, or converted after extraction (before, on, or after the date of enactment of section 651(e) of the Energy Policy Act of 2005), for use for a commercial, medical, or research activity.

any state, from requiring a licence to receive, possess, use or transfer unimportant quantities of source material.

(b) If the material in question contains uranium and/or thorium at a concentration of 0.05% by weight or more, in any physical or chemical form, then it is 'licensable' source material, and its receipt, possession, use and transfer are subject to a general or specific licence issued by NRC or the relevant Agreement State.

(c) If the material in question is an 'ore' that is above 0.05% by weight uranium and/or thorium, then it is source material in its entirety.[20] As long as it is 'unrefined or unprocessed ore',[21] it is exempt from licensing under 10 CFR 40.13(b). However, any source material processing triggers the requirement for a specific licence. [NRC Health Physics Position (HPPOS) No. 184 explains that "any processing or refining may alter the radiological environment associated with the source material enough so that the health and safety of workers and others becomes a matter of legitimate regulatory concern."]

(d) With respect to ore, the NRC commented: "There is no consistent definition of 'ore' underlying the regulatory decisions made by the Commission. The word 'ore' is not defined in Part 40 or in the AEA of 1954, as amended. Under the present regulatory scheme, and depending on the interpretation of the word 'ore,', materials with low concentrations of uranium and thorium could be considered AEA material exempt from regulation or might not be considered AEA material at all. The only formal Agency definition of 'ore' is in guidance for use only with respect to alternate feed material. This guidance defines 'ore' in terms of its use for processing for its uranium or thorium content at a uranium recovery facility. This lack of a consistent definition of ore contributes to problems with the interpretation of the definition of source material, which have resulted in numerous regulatory and legal deliberations and inconsistencies in the regulation of source material."[22]

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[20] In the uranium milling context, NRC's alternate feed guidance defines ore as: "Ore is a natural or native matter that may be mined and treated for the extraction of any of its constituents or any other matter from which source material is extracted in a licensed uranium or thorium mill." 60 Fed. Reg. 49296, September 22, 1995.

[21] Neither the AEA nor NRC regulations define 'ore.' 'Unrefined and unprocessed ore' is defined at 10 CFR 40.4 as "means ore in its natural form prior to any processing, such as grinding, roasting or beneficiating, or refining".

[22] SECY-03-0068.

What, then, is TENORM? There are two competing definitions of TENORM that have found their way into state — but not federal — regulations. One definition of TENORM that is endorsed by EPA is that used by the National Academy of Sciences, Commission on Life Sciences in its Evaluation of Guidelines for Exposure to Technologically Enhanced Naturally Occurring Radioactive Material (National Academy Press, 1999):

> "Technologically enhanced naturally occurring radioactive materials are any naturally occurring radioactive material not subject to regulation under the Atomic Energy Act whose radionuclide concentrations or potential for human exposure have been increased above levels encountered in the natural state by human activities."

A different definition of TENORM is that of the Conference of Radiation Control Program Directors, Inc. (CRCPD), a non-governmental, non-profit professional organization founded "to promote consistency in addressing and resolving radiation protection issues, to encourage high standards of quality in radiation protection programs, and to provide leadership in radiation safety and education".[23] Part N of the CRCPD's Suggested State Regulations for the Control of Radiation (SSRCRs) addresses regulation and licensing of TENORM. Section N.3 defines TENORM as:

> "Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) means naturally occurring radioactive material whose radionuclide concentrations are increased by or as a result of past or present human practices. TENORM does not include background radiation or the natural radioactivity of rocks or soils. TENORM does not include 'source material' and 'byproduct material' as both are defined in the Atomic Energy Act of 1954, as amended (AEA 42 USC §2011 *et seq.*) and relevant regulations implemented by the NRC."

It is immediately apparent that the more expansive EPA-supported definition of TENORM requires *either* an increase in radionuclide concentration over its natural state *or* an alteration in potential exposure pathways of an un-concentrated natural material for it to be TENORM. CRCPD, on the other hand, requires a material's radionuclides to become concentrated over its natural state to become TENORM. Both definitions exclude AEA materials (as they must under AEA preemption discussed above), but phrase it differently: The EPA-

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[23] See www.crcpd.org.

supported TENORM definition *includes* materials *not subject to regulation* by the AEA, while CRCPD's definition expressly *excludes* 'source material' and 'byproduct material' as defined by the AEA and NRC regulations. Under CRCPD's TENORM definition, materials that have been merely physically relocated in the environment and do not exhibit a quantitative increase in radionuclide content are excluded from the definition. Consider radionuclides in uranium or phosphate mine spoils that became relocated to the surface. Under the EPA-supported TENORM definition, this material is TENORM; under CRCPD's definition it is not.

One might ask whether the exclusionary phrase 'subject to regulation' under the AEA requires active regulation, such as licensing or other affirmative control, in order for a material to be excluded from the EPA supported TENORM definition. Could this phrase be interpreted to mean that materials not subject to licensing — like unimportant quantities of source material — are not subject to regulation and therefore open to regulation as TENORM? Unimportant quantities below 0.05% uranium and thorium and unrefined/unprocessed ore *are* subject to the AEA's regulatory jurisdiction, but the regulatory body responsible for administering the AEA — the NRC — has made an affirmative decision through rulemaking to exempt these materials from the requirement for licensing.

It is noteworthy that at the time of this writing, TENORM is not defined anywhere in any federal statute or federal agency regulation. TENORM and its role in source material licensing was, however, recently considered by the Nuclear Regulatory Commission in *In the Matter of Hydro Resources, Inc.*, CLI-06-14 (2006) ('HRI'). The case involved an appeal to the Commission of a challenge to HRI's *in situ* uranium mining licence because NRC did not consider potential doses arising from uranium bearing mining spoil left on-site by the site's previous owner to be a factor in determining 'public dose' for licensing purposes. The Commission's decision upholding the Presiding Officer's rejection of the Intervenor's challenge provides a useful discussion of NORM, TENORM and 'background' insofar as NRC licensing decisions are made:

> In 1991, NRC published revisions to the standards in 10 C.F.R. Part 20 for protection against radiation. In its definition of 'background radiation', the rule expanded the category of what was once called 'natural background' radiation to include various anthropogenic sources as well as NORM, and to expressly exclude NRC-regulated sources:
>
> Radiation from cosmic sources; naturally occurring radioactive material, including radon (except as a decay product of source or special nuclear material); and global fallout as it exists in the environment from the testing of nuclear explosive devices or from past nuclear accidents such as

> Chernobyl that contribute to background radiation and are not under the control of the licensee. ‘Background radiation’ does not include radiation from source, byproduct, or special nuclear materials regulated by the Commission.

‘Naturally occurring radioactive material’ — NORM — is not defined elsewhere in the regulations.

The Presiding Officer addressed the last sentence of the ‘background radiation’ definition first, concluding that radiation from ‘source material’ can be background radiation where, as here, the source material from which it emanates is not ‘source material . . . regulated by the Commission’. While the Atomic Energy Act provides that uranium and thorium are source material, the Presiding Officer explained, the NRC does not regulate *all* source material. Unprocessed ores and source material with insignificant concentrations of radionuclides are not regulated by the Commission. Because mining spoil is unprocessed ore and thus *not* ‘regulated by the Commission’, the Presiding Officer determined that the last sentence of the definition did not preclude his finding that the radiation from mining spoil constituted ‘background radiation’. The Presiding Officer went on to conclude that mining spoil should be considered NORM, and thus background radiation within the first sentence of our definition. He found that the mining spoil falls within the scope of TENORM.

The Presiding Officer's understanding of our "background radiation’ definition is correct. At the time the NRC drafted the regulation defining ‘background radiation,’ the term NORM was understood to include TENORM. This is evident from the definition's history. It shows that the NRC considered, and explicitly rejected, a suggestion by the Advisory Committee on Reactor Safeguards (‘ACRS’) that the proposed rule be revised to ‘emphasize’ that NORM did *not* include TENORM. As the ACRS suggestion implicitly recognized, *excluding* TENORM would have required express language, if that was what the NRC had intended with this regulation. But the agency rejected the ACRS suggestion, for the reason that most TENORM is outside NRC's regulatory jurisdiction. As the Presiding Officer recounts, over the years the NRC and other regulatory authorities have repeatedly considered ‘TENORM’ as equivalent to ‘NORM’.

* * *

Intervenors are simply mistaken in their assertion that TENORM only designates materials, such as plasterboard and fertilizer that have been manufactured for a use unrelated to their incidental radioactive properties. The EPA, which regulates TENORM, describes TENORM as including waste streams from various industries, such as sewage treatment waste and waste from drinking water treatment. Consistent with this, the NRC has recognized that TENORM includes waste materials:

> TENORM is found in various concentrations in a variety of forms (physical and chemical matrices) such as scrap metal, sludges, fluids, scales in storage tanks and piping, chemical residues, processing fluids, surface and groundwaters, and mine tailings.
>
> As shown above, the understanding at the time the regulation issued implicitly included TENORM as a type of NORM. The fact that NRC regulations do not define 'TENORM,' as such, is not surprising. There is no need for the NRC to draw fine distinctions among various classes of materials that it does not even regulate; the spoil leftover from mining falls into that category."[24]

The HRI case teaches that 'source material' can be background radiation where … the source material from which it emanates is not 'source material...regulated by the Commission' and that source material not regulated by NRC is TENORM, which is considered equivalent to NORM and dose from TENORM is 'background', excluded from consideration in licensing decisions. But if the uranium mining spoil — an unrefined and unprocessed ore — is exempt from regulation by NRC, does this mean that some other authority, such as a state or different federal agency, can assert its own regulatory jurisdiction over uranium mine spoils or any other unimportant quantity 'not regulated by the Commission'? Could a State regulate uranium mine spoil as TENORM?

Uranium mine spoils (unprocessed ore) and other unimportant quantities (such as materials below 0.05% U and /or Th) *contain* source material under NRC's *exclusive* jurisdiction. NRC has grappled with this issue for years: because unimportant quantities (TENORM) are not part of NRC's core mission — regulating the nuclear fuel cycle — and regulating TENORM would expand the Commission's regulatory reach into activities far removed from its core mission, how can the NRC limit its own jurisdiction over source material so that other federal agencies (or States) could then regulate these materials? Beginning in 1999, NRC began to search for a resolution of how the Commission could

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[24] *In the Matter of Hydro Resources, Inc.* CLI-06-14 (2006) at 4-5 (citations omitted).

delegate or transfer its jurisdiction over unimportant quantities to other regulatory entities, and the NRC Staff wrote:[25]

> "Based on the mission of the NRC under the AEA, it could be argued that NRC authority should be limited to activities related to recovery of thorium and uranium (primarily in support of the nuclear fuel cycle). . . . Naturally occurring uranium and thorium inadvertently concentrated by various other processes might more easily and consistently be regulated along with other naturally occurring materials (by EPA, Occupational Safety and Health Agency (OSHA), and the States).
>
> Among the various options . . . involve NRC relinquishing authority for some low-level source material to these other agencies. . . .
>
> The [NRC] staff believes that a legislative option could fully resolve the issues related to this exemption. The most appropriate might be to limit NRC authority to activities related to recovery of uranium and thorium (primarily in support of the nuclear fuel cycle)."

Thus the Interagency Jurisdictional Working Group (JWG) was created in 2000, comprising NRC, EPA, OSHA and the States (including the Organization of Agreement States (OAS) and CRCPD), with representatives from other federal agencies (including DOE, Army Corps of Engineers, DOT and Department of Interior). In 2003, the JWG concluded:

> "Based on its evaluations, the general consensus of the JWG is that the best approach for regulating low-level source material is to limit NRC authority to uranium and thorium that are extracted (i.e. purposely concentrated for the use of the uranium or thorium). Once extracted/purposely concentrated, the uranium and thorium would continue to be considered source material subject to NRC regulations. All other incidental uranium and thorium that are not extracted or purposely concentrated would be considered NORM/TENORM, and would be regulated by current standards/regulations for this material, under the regulatory programs of other agencies, such as EPA, OSHA, and the States, to the extent that these organizations choose to regulate this material."[26]

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[25] SECY 99-259 Exemption in 10 CFR Part 40 for materials less than 0.05 per cent source material – optios and other issues concerning the control of source material.

[26] SECY-03-0068 at page 8.

The JWG faded into the background for the next three years, but interest was rekindled at the Commission level in *Termination of Nuclear Regulatory Commission license for the Heritage Minerals, Inc. site in Manchester Township, New Jersey.*[27] Heritage Minerals, Inc. (HMI) operated a mineral sand processing facility that used gravimetric and electrostatic techniques to separate zircon and titanium ores (ilmenite and rutile) from unwanted ‘gangue’ containing silica sand and the mineral monazite, which contains uranium and thorium above 0.05%. Under normal operating conditions the monazite was not intended to be separated from the gangue, and was therefore kept below 0.05% uranium and thorium. Under ‘upset’ conditions, when a processing stage became inoperative, plant operators would divert the monazite stream from the process in order to keep the machinery running. It was these on-site accumulations of monazite that resulted in the NRC licence.

> “The HMI site is a former minerals mining and processing facility, owned by HMI, which is located in Manchester Township, New Jersey within the Pine Barrens. The site, like the surrounding properties in Ocean County is flat with coastal sands. Although the site consists of almost 7000 acres, mining and processing operations took place on approximately 287 acres, and NRC licensed areas comprised less than one acre. The facility was used from 1973-1989 for the mechanical processing of dredged native sand to extract titanium- and zirconium-bearing heavy minerals. The native sand also contains natural uranium and thorium [in the form of monazite], which were concentrated in the process waste tailings. In 1989, HMI began reprocessing the tailings to extract any remaining heavy minerals. The resultant waste tailings contained a concentration of uranium and thorium in excess of 0.05% by weight, exceeding the 10 CFR 40.13(a) unimportant quantity exemption for source material. HMI segregated the source material and applied for an NRC licence.
>
> Before the licence was issued, reduced demand caused HMI to cease processing activities. The NRC licence, issued in 1991, authorized possession of the stockpiled source material and decommissioning of the impacted areas of the site (specified as the two mill buildings and the ground beneath the stockpile).”[28]

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[27] SECY-06-0117.

[28] SECY-06-0117.

In addition to the NRC licensed monazite concentrations, there existed some areas of elevated uranium and thorium, but since concentrations were below 0.05%, NRC excluded these areas from its licensing jurisdiction:

> "The ground between and surrounding the impacted areas contains diffuse thorium and uranium concentrations above background but below 0.05%, resulting from staging and regrading waste sands from previous (unlicensed) processing activities. This material remained exempt from NRC regulations."[29]

It was the fate of the non-NRC licensed areas of TENORM contamination that caused trouble: NRC considered, but declined, to address TENORM decontamination under the site licence. New Jersey, on the other hand, pressed NRC to clean up the entire site, notwithstanding the Commission's lack of jurisdiction over TENORM:

> "The [NRC] staff dose assessment only considered the exposure to the public from the residual radioactivity within the boundary of the NRC licensed areas. The average concentration of thorium-232 (the greatest contributor to radiation exposure) within this area is 2.3 pCi/g [0.085 Bq/g]. The soil outside the NRC licensed areas contains measured thorium concentrations ranging from well below 1 pCi/g [0.037 Bq/g] to greater than 44 pCi/g [1.63 Bq/g]. Source material concentrations outside the NRC licensed areas may exist up to just below 0.05%. The dose to the public from the elevated concentrations of natural thorium and natural uranium from past site operations is expected to exceed the calculated dose within the licensed area. Because this material originated from unlicensed activities, and never reached the defined concentration of source material, NRC did not require its remediation. Decommissioning of the remainder of the site falls under the jurisdiction of the State of New Jersey. HMI has provided the New Jersey Department of Environmental Protection (NJDEP) its proposed plan for remediation of the entire site to the state cleanup levels, based on 15 mrem/year [0.15 mSv/y] TEDE."[30]

In the Commission Voting Record on SECY-06-0117 approving HMI's licence termination, Commissioner Jaczko (now NRC Chairman) expressed his own confusion over NRC's jurisdictional limitations on TENORM and suggested

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[29] *Id.*

[30] SECY-06-0117.

the Commission consider *lowering* the 0.05% exemption in 40.13(a) to capture such materials:

> “With regard to the remediated areas, I am concerned by the inconsistent, complex and confusing regulatory and legal framework governing the decommissioning — and even licensing and operation — of sites containing uranium and thorium in concentrations below 0.05% by weight (the regulatory threshold for description of uranium and thorium as ‘source material’) *and* in conditions distinct from a natural state. In this case, the staff correctly interpreted commission regulations and orders that consider this uranium and thorium to be technologically enhanced naturally occurring radioactive material (TENORM), which Commission regulations and precedents consider a subset of naturally occurring radioactive material (NORM). NORM classes of uranium and thorium are not regulated by the Commission and are excluded by regulation from consideration when analyzing the sources of radioactive materials that could contribute doses to the public and workers.
>
> I am troubled by this situation, because uranium and thorium classified as TENORM often has significant dose implications for the public. At the Heritage site there are some locations where the uranium and thorium material falls below the source material threshold by only a small amount. The threshold for determination of source material is not based on the expected dose contributions from material below this threshold, but from the ability of the uranium and thorium to be useful for commercial nuclear purposes. In many areas at the Heritage site, there could be doses on the order of hundreds of millirem from the uranium and thorium material classified as TENORM.
>
> In fact at the Heritage site, the NRC has required the clean-up of areas containing licensed uranium and thorium source material, but the NRC cannot require the clean-up of areas containing unlicensed uranium and thorium TENORM material directly adjacent to areas containing unlicensed material. In some instances, the dose implications from the licensed and unlicensed materials are comparable. As a result, there are pockets of decontaminated areas in a larger region that still contains significant areas of contamination.
>
> Unfortunately, the arcane nature of the Commission's regulations prevents this material from being properly decommissioned by the NRC. The responsibility for ensuring the further decontamination resides with the

> Environmental Protection Agency and the states. I do not find this an acceptable alternative, but without a rulemaking initiative or an order, the Commission is unable to impose additional decommissioning requirements on the Heritage site. To address this, the Commission should consider revising the definition of source material to consider a threshold for uranium and thorium concentrations that is based on the public health and safety dose implications of the material."[31]

NRC Commissioner McGaffigan, with supporting supplemental comments from then NRC Chairman Klein and Commissioners Merrifield and Lyons, suggested that instead of increasing NRC regulatory oversight of TENORM, NRC should accelerate efforts of the JWG's to limit NRC's authority over unimportant quantities:

> "In his vote, Commissioner Jaczko supports the tightening of NRC regulatory control over radioactive material containing uranium and thorium. Among other things, he proposes that the Commission revise "the definition of source material to consider a threshold for uranium and thorium concentrations that is based on public health and safety dose implications of the material." This revision would require that we lower the concentration limit of 0.05 weight per cent in 10 CFR 40.13(a). This action would only further exacerbate the existing problem of dual-regulation of technologically-enhanced, naturally-occurring radioactive material (TENORM). The Commission, the NRC staff and our partners in other Federal agencies and the States have been deliberating on this matter for years . . . , and SECY-03-0068). By lowering this concentration, NRC would add untold number of other industries and activities (such as coal ash, petroleum extraction, etc.) to those that already fall under our authority, with no additional regulatory benefit beyond that already provided by oversight by the EPA, OSHA, and the States.
>
> I propose an alternative solution. . . . to seek a legislative change to limit NRC authority to uranium or thorium that are extracted or purposely concentrated for the use of uranium or thorium."[32]

The current status of the JWG's efforts to divest NRC of its jurisdiction over uranium/thorium that is not 'purposely concentrated' (which presumably

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[31] SECY-06-0117 CVR, Comments of NRC Commissioner (now Chairman) Jaczko.

[32] SECY-06-0117, Comments of NRC Commissioner McGaffigan.

includes most unimportant quantity TENORM) is not clear. It is anticipated that a legislative proposal to limit NRC's jurisdiction over source material may be released in mid-2010. One thing is certain: NRC cannot simply delegate or transfer its AEA jurisdiction over unimportant quantity TENORM to other agencies or the states through rulemaking or by a Memorandum of Understanding (MOU). Congress vested the Commission with exclusive jurisdiction over all source material, including those quantities deemed unimportant. It is only Congress – through legislative amendment of the AEA — that can change the Commission's jurisdiction, or provide a mechanism for NRC to delegate limited jurisdiction to others. It is also fairly certain that until the AEA is amended there will be continuing uncertainty over whether other federal agencies or the states can regulate TENORM that is an unimportant quantity of source material.

2. PRACTICAL ASPECTS OF STATE TENORM REGULATION

A few States have NORM or TENORM regulations that expressly define the types of materials subject to regulation, provide for exemption of certain materials, and establish thresholds for regulation based on radionuclide content. Most, but not all, State NORM/TENORM regulations establish a regulatory threshold based on the radium content of a material (total Ra-226 and Ra-228). Radium that is not a 'discrete source of Ra-226' covered by the new 11.e.3 byproduct material definition remains outside NRC's AEA jurisdiction, and is therefore amenable to regulation by the States. Some of the States that have adopted NORM/TENORM regulations follow the CRCPD Part N model TENORM rule and establish a regulatory threshold at 5 pCi/g (0.187 Bq/g) Ra-226 and Ra-228. The following discussion addresses a few practical examples of State TENORM regulation and how potentially regulated entities might be confronted with TENORM issues arising under State authority.

An interesting question arises in the context of state TENORM regulation: States that regulate TENORM based on its radium content typically assert that State regulatory jurisdiction attaches only to the Ra-226 (or in the case of Th-232, Ra-228) that is present in the natural uranium decay series. No jurisdiction is asserted over the U-238 or Th-232 parent isotope, just the radium progeny. In cases where Ra-226 /-228 have partitioned to certain media (e.g. scales seen on process equipment), the situation is easy: no federal AEA preemption is in question and the State has jurisdiction over the radium progeny when it exists in physically separate form from its source material parents. On the other hand, where natural U-238 and Th-232 exist in equilibrium with their radium decay progeny, as in unrefined or unprocessed ore and many processed minerals and residues, the issue is more difficult: Where the radium progeny are physically co-located with their source material parent isotopes in natural ratios, can a State assert TENORM licensing jurisdiction over the radium alone without running afoul of the AEA's preemptive prohibition against licensing unimportant quantities? Under such conditions the State's imposition of a TENORM licence on radium contained in an unimportant quantity means that the entire unimportant quantity is licensed by the State. This seems to contradict the AEA Section 62 prohibition against licensing unimportant quantities. This issue has not yet been resolved in any administrative proceeding or judicial forum, but it did arise in the following administrative appeal of a State TENORM licensing decision.

2.1. The TENORM case in State A

2.1.1. Background

Company owns a facility in the State of A that formerly processed titanium ores by separating iron oxide (IOX) which was then deposited in a surface impoundment. In 2005 it became apparent that resurgent market conditions for raw materials had made IOX a valuable commodity that could be recycled as iron ore.

Data obtained by the Company and provided to the State Radiation Control authority showed that IOX contains uranium up to 11 parts-per-million (ppm) (0.0011 %) and thorium up to 256 ppm (0.0256%), along with the radium-226 and radium-228 isotopes at equilibrium concentrations that are naturally affiliated with uranium and thorium.

The State Radiation Control authority took the position that because IOX contained greater than 5 pCi/g [0.185 Bq/g] radium-226 and radium-228, it was TENORM and the Company would be required to apply for a specific TENORM licence for possession or disposal.

The Company filed an administrative appeal of the regulatory body's decision.

Two arguments were raised in the appeal: (1) the State has not complied with its Administrative Procedures Act by failing to promulgate any regulation defining TENORM and the conditions under which TENORM is licensed and (2) the State is preempted from licensing unimportant quantities of source material. These arguments are addressed in turn.

2.1.2. The State Administrative Procedures Act

The Code of State A enumerates the State Radiation Control Agency's duties, including the duty to:

> “*Formulate, adopt, promulgate and repeal codes, rules and regulations* relating to control of sources of ionizing radiation with due regard for compatibility with the regulatory programs of the federal government.”

State A’s radiation control regulations do not include a definition of ‘TENORM’. Nowhere in the regulations can be found any standard establishing the “5 pCi/gm threshold above which a specific licence is required” as asserted by the Office of Radiation Control.

The Office of Radiation Control’s NORM web page, on the other hand, explains the criteria ‘adopted’ by ORC for identifying licensable NORM:

> “To determine if the NORM concentration is such that it will require a radioactive material licence, the Alabama Office of Radiation Control has adopted two thresholds. The first threshold is an exposure reading of 50 microroentgen/hour (μR/h) (background included) at contact with the NORM or NORM contaminated article. This threshold is only to be used for discreet [sic] items such as pipes or tanks, and the TENORM contaminated scale or sludge contained in these pipes or tanks. The second threshold is a concentration of greater than 5 picocuries/gram (pCi/g) of radium 226. If either of these thresholds are exceeded, a radioactive material licence is required to receive, possess, use, transfer, own or acquire the NORM.”

Significantly, the 5 pCi/g [0.185 Bq/g], the 50 μR/h criteria and their role in categorizing NORM or TENORM as licensable or exempt from licensing do not appear in any of the Office of Radiation Control’s licensing regulations.

Every State government has enacted statutory requirements implementing procedures which must be followed by State agencies. The Administrative Procedures Act of State A prescribes the procedures that must be followed in any State Agency rulemaking, and defines a ‘rule’ as:

> “RULE. Each agency regulation, standard, or statement of general applicability that implements, interprets, or prescribes law or policy, or that describes the organization, procedure, or practice requirements of any agency…”

The Office of Radiation Control’s assertion that 5 pCi/g [0.185 Bq/g] radium-226 and radium-228 is a licensing criterion is a ‘standard or statement of general applicability’ and is therefore a ‘rule’.

In invalidating an Agency directive that was improperly promulgated as a ‘rule’ the State A Supreme Court held:

> "The provisions of the Administrative Procedure Act impose upon administrative agencies the duty—preliminary to the 'adoption, amendment, or repeal,' – to publish the 'terms or substance' of such rules, and, among other things, to 'afford all interested persons reasonable opportunity to submit data, views, or arguments, orally or in writing.' Noncompliance with these provisions voids every 'agency rule, order, or decision' taken in any case in which the provisions are applicable."[33]

Publishing a TENORM definition and licensing policy on an Agency Internet web page is *not* the equivalent of publishing a proposed rule for public comment as prescribed by the Administrative Procedures Act.

2.1.3. Federal AEA preemption under 10 CFR 40.13(a)

State A, as an 'NRC Agreement State' has promulgated regulations defining 'source material' and implementing source material licensing requirements that are compatible with those of NRC.[34]

Since the radionuclide content of IOX is predominantly thorium, with some uranium present, the IOX contains 'source material'.

Congress excluded certain concentrations of source material from NRC's licensing authority. Section 62 of the AEA expressly states that "*licences shall not be required* for quantities of source material which, in the opinion of the Commission, are unimportant."[35]

In accordance with Congress' express mandate in Section 62 of the AEA, NRC regulations provide a definition of 'unimportant quantities' of 'source material', including:

> "source material in *any chemical mixture, compound, solution, or alloy* in which the source material is by weight less than one-twentieth of 1 per cent (0.05 per cent) of the mixture, compound, solution or alloy. …"[36]

Analysis of IOX confirms that the material is below 0.05 weight per cent uranium and thorium. IOX is therefore an unimportant quantity of source material under federal NRC and corresponding State A regulations, and the

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[33] *Brunson Construction & Environmental Services, Inc. v. City of Prichard*, 664 So. 2d 885, 893 (Ala. 1995).

[34] Please see Footnote 13 for an overview of NRC Agreement States.

[35] 42 U.S.C. 2092 (emphasis added).

[36] 10 CFR § 40.13(a)(emphasis added).

receipt, possession, use, or transfer of IOX is therefore exempt from licensing. More detailed analysis of the uranium, thorium and their decay progeny, including radium-226 and radium-228, present in IOX confirms that source material in IOX is present in *equilibrium.* That is, the uranium-238 and thorium-232 *parent* isotopes exist along with their respective radium-226 and radium-228 *progeny* in concentrations that are representative of, and predicted by, the natural and predictable radioactive decay of uranium and thorium over time. In other words, neither uranium nor thorium present in IOX had been altered from a composition that is found in nature. Therefore, since the IOX material at the Company's facility is an unimportant quantity of source material that does not exceed one 20th of one per cent source material, by weight, the IOX should be excluded from licensing as an unimportant quantity of source material.

The State asserted that even if unimportant quantity source material was exempt from licensing, it could still be subject to licensing as NORM or TENORM if the radium-226 and radium-228 *contained in* the source material exceeded the 5 pCi/g [0.185 Bq/g] licensing threshold. This assertion is contrary to the express prohibition of Section 62 of the AEA against any licensing of ‘unimportant quantities’ of source material.

Reading AEA Section 62 as prohibiting licensing of the uranium-238 and thorium-232 *parent* isotopes in unimportant quantity source material, while at the same time allowing licensing of the radium-226 and radium-228 *progeny* that naturally coexist in unimportant quantities of source material would render AEA Section 62 a nullity. In particular, where the AEA defines source material to include uranium and thorium “in any physical or chemical form”, a logical conclusion is that this definition applies to the form of uranium and thorium that found in nature, and in equilibrium with its radium decay progeny.

2.1.4. The outcome

After extensive negotiations, this case settled. It is unknown whether the above arguments would have prevailed before an agency tribunal or a reviewing court. The terms of the settlement were favourable to both sides: the Company's IOX was exempt from TENORM licensing, provided it was below 1 Bq/g and exported from the State of A. The Company agreed to apply for a TENORM licence which covered possession, decontamination and disposal of radium scales found in process equipment during dismantling of the facility. The Company also agreed to implement a comprehensive NORM Awareness Programme which provided for worker education, training, and dose monitoring, as necessary to comply with State A's radiation protection standards.

2.2. TENORM and CERCLA in *Amoco v. Borden*

We turn now to the most important federal environmental statute that addresses environmental remediation: the Comprehensive Environmental Response, Compensation and Liability Act ('CERCLA' or 'Superfund').[37] CERCLA is founded on the 'polluter pays' principle and imposes strict, joint and several and *retroactive* liability on 'potentially responsible parties' (PRPs) that impair the environment by causing the 'release or threatened release' of 'hazardous substances.' In addition to liability provisions, CERCLA has cost-recovery provisions under which PRPs are required to reimburse the federal government (EPA) or private parties for response costs incurred in relation to the release of 'hazardous substances'.

The case of *Amoco v. Borden*[38] is an appellate decision involving a private cost recovery action for environmental cleanup costs incurred by the buyer (Amoco) against the seller (Borden) of property that was found to be contaminated with phosphogypsum residue containing TENORM.

> "The property at issue is a 114 acre tract of land in Texas City, Texas. For many years, Borden operated a phosphate fertilizer plant on the site. As a by-product of the fertilizer manufacturing process, large quantities of phosphogypsum were produced. The site now contains a large inactive pile of phosphogypsum covering approximately 35 acres.
>
> Phosphogypsum alone contains low levels of radioactivity. More highly radioactive sludges and scales from processing equipment, however, were dumped into the phosphogypsum pile, creating 'hot' areas within the pile. Additionally, during processing, radioactive materials became concentrated in manufacturing equipment, pipe, and filter cloths used in production. These materials constitute 'off-pile' wastes and were left primarily near a junkyard on the property and near the abandoned manufacturing buildings. Some of the off-pile sites contain over 500 times the background level of radiation."

In rejecting Amoco's CERCLA claim, the trial court held that Amoco must prove that some threshold level of radioactivity exists at the site in order to establish CERCLA liability and selected the standards for remedial actions at inactive uranium processing sites [5 pCi/g [0.185 Bq/g] radium averaged over the

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[37] 42 USC 103, *et seq.*

[38] 889 F.2d 664 (5th Cir. 1989).

first 15 cm of soil below the surface, and 15 pCi/g [0.555 Bq/g] radium averaged over 15 cm thick layers of soil more than 15 cm below the surface].[39]

The trial court concluded that contamination did not exceed this standard and entered judgment for Borden. Amoco appealed the court's decision that a threshold level of radionuclides must be shown to exist at the site to establish CERCLA liability, the appropriateness of the Inactive Tailings Standards for defining that threshold, and the court's application of that standard.

The appeals court found that seller's (Borden's) facility fell within the statutory framework of CERCLA, but liability and cost recovery would depend on whether a 'hazardous substance' had been released at the site: The court next considered whether Ra-226 in phosphogypsum is a CERCLA 'hazardous substance':

> "Radium-226, the primary radioactive waste on the property, decays to form a gas, radon-222, and solid 'daughter products.' Radon and its daughter products are considered radionuclides, which are defined as "any nuclide that emits radiation." 40 C.F.R. Sec. 61.91(c) (1988). The term hazardous substance includes "any element, compound, mixture, solution, or substance designated pursuant to section 9602 of [CERCLA], ... [and] any hazardous air pollutant listed under section 112 of the Clean Air Act...." Sec. 9601(14). The EPA has designated radionuclides as hazardous substances under Sec. 9602(a) of CERCLA. See 40 C.F.R. Sec. 302.4 (1988). Additionally, the regulations promulgated by the EPA under Sec. 112 of the Clean Air Act, 42 U.S.C. Sec. 7412, list radionuclides as a hazardous air pollutant. See 40 C.F.R. Sec. 61.01(a) (1988)."

Importantly, the court concluded that there is no *quantitative* limitation on the amount of Ra-226 present in order to find that a 'release' of a CERCLA 'hazardous substance' has taken place:

> "The plain statutory language fails to impose any quantitative requirement on the term hazardous substance and we decline to imply that any is necessary. Radionuclides meet the listing requirements and therefore the radioactive materials on Amoco's property are hazardous substances within the meaning of CERCLA."

The 5th Circuit decision in *Amoco v. Borden* stands for the proposition that *all* radionuclides are encompassed by the CERCLA definition of 'hazardous

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[39] 40 C.F.R. Part 192 (1988) ('Inactive Tailings Standards').

substances' because radionuclides are identified, without any limitation, as hazardous air pollutants under Section 112 of the Clean Air Act. The court found that no quantitative limitation applied to the release of Ra-226 in order to find that a 'release' of a hazardous substance occurred. This case serves as important precedent in public (EPA) and private cost recovery actions under CERCLA.

2.3. California Proposition 65

Not all statutes that implicate TENORM involve licensing or remedial actions. The California Safe Drinking Water and Toxics Enforcement Act of 1986 was enacted as a ballot measure ('Proposition 65') intended to protect Californians against exposure to "substances known to the State of California" to cause cancer or reproductive harm.

> "No person in the course of doing business shall knowingly and intentionally expose any individual to a chemical known to the state to cause cancer or reproductive toxicity without first giving clear and reasonable warning to such individual…"[40]

Proposition 65 requires the Governor to annually publish a list of chemicals known to the State of California to cause cancer or reproductive toxicity:

> "List of Chemicals Known to Cause Cancer or Reproductive Toxicity. (a) On or before March 1, 1987, the Governor shall cause to be published a list of those chemicals known to the state to cause cancer or reproductive toxicity within the meaning of this chapter, and he shall cause such list to be revised and republished in light of additional knowledge at least once per year thereafter…"[41]

Among the many chemical substances on the current (February 4, 2010) Proposition 65 list are: 'radionuclides' and 'thorium dioxide', both being listed as 'chemicals known to the State of California' to cause cancer.

The Proposition 65 enforcement provisions are steep; but most significant is that in addition to the California Attorney General, private individuals may bring action to enforce Proposition 65 'in the public interest' and collect attorney's fees for their efforts. Also, an individual need not show any particular exposure or harm from the listed substance in order to prevail: all that is needed is

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[40] Cal. Health & Safety Code Sect. 25249.6.

[41] Cal. Health & Safety Code Sect. 25249.8.

to prove (1) the product is sold in the State of California; (2) the product contains a listed substance; and (3) the product does not bear a ‘clear and reasonable warning’ that it contains a listed substance.

Proposition 65 might be considered a statute with good intentions that has been hijacked by ‘bounty hunters’: since its passage, a multitude of Proposition 65 plaintiff’s lawyers have been targeting products sold in California without a ‘clear and reasonable warning’ that the product contains a listed substance (some examples: lead in brass faucets; 1,1,1-trichloroethane in ‘white out’ correction liquid; crystalline silica in mineral products).

Does Proposition 65 apply to TENORM? Yes, although its applicability to AEA materials would very likely be challenged under *Pacific Gas & Electric*, discussed earlier. However, there are as yet no reported Proposition 65 decisions alleging failure to warn of the presence of radionuclides in a product. Given the ubiquitous presence of radionuclides in everything on earth, this is a pretty broad reach. Is there a threshold below which the radionuclide content of a product? One exemption to the Proposition 65 warning requirement is:

> “An exposure for which the person responsible can show that the exposure poses no significant risk assuming lifetime exposure at the level in question for substances known to the state to cause cancer, and that the exposure will have no observable effect assuming exposure at one thousand (1000) times the level in question for substances known to the state to cause reproductive toxicity, based on evidence and standards of comparable scientific validity to the evidence and standards which form the scientific basis for the listing of such chemical pursuant to subdivision (a) of Section 25249.8. In any action brought to enforce Section 25249.6, the burden of showing that an exposure meets the criteria of this subdivision shall be on the defendant.”

Therefore, a defendant in a Proposition 65 suit alleging failure to warn of the presence of radionuclides would have to present evidence to show that the level of exposure to radionuclides posed ‘no significant risk assuming lifetime exposure at the level in question’. Given the current debate over radiation risk, and EPA’s recent radiogenic risk models[42], the arguments for the defense might be interesting and would certainly involve some compelling testimony by the health physics expert.

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[42] EPA Radiogenic Cancer Risk Models and Projections for the US Population (Draft) (December 2008).

3. CONCLUSIONS

This paper was written with the objectives of providing an overview of the complex jurisdictional issues that pervade the regulation of TENORM in the USA legal system. It is hoped that the following take home questions are remembered whenever a TENORM situation is encountered: Which regulatory body has the legal authority to regulate TENORM? Is the regulation of the particular TENORM in question preempted by the AEA? Has the regulatory body (state or federal) complied with the Administrative Procedures statute governing its action in regulating TENORM? And finally, what regulatory action is being sought: licensing for possession, use, or transfer; remediation of contamination; or is there some other health and safety aspect involved (as in California Proposition 65)?

For the time being, the uncertainty with respect to US TENORM regulation will remain the *status quo*. Perhaps someday a uniform, harmonized, regulatory scheme will exist that affords regulatory certainty to persons engaged in the generation, use, and disposal of TENORM. Until that time, we must continue to work within the existing jurisdictional framework governing the naturally occurring radionuclides that comprise TENORM.

DEVELOPMENT OF NORM MANAGEMENT IN AUSTRALIA

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Abstract

Australia has had a long involvement with NORM, mainly because of mining and processing of mineral ores. Radium mining was carried out in the early 20th century. After 1949, there was a rapid expansion in uranium mining (particularly in the Northern Territory) and mineral sand mining. Australia is a Federation of the Commonwealth, six States and two Territories, which independently regulate within their jurisdiction. Early mineral extraction and processing operations were unregulated, resulting in the existence of a large number of legacy sites. Regulation of uranium mining started in the 1970s, and mineral sand mining after 1980. The regulations vary in detail between jurisdictions. Most other NORM situations are not regulated in any of the States or Territories. A major review of the Western Australian mineral sand industry in the 1980s led to considerable reductions in doses to workers. Remediation of many of the old uranium mine sites in the Northern Territory has been carried out over the last 15 years and is continuing. International awareness of NORM as a potential source of risk to workers, members of the public and the environment has increased significantly in recent years. After an extensive stakeholder consultation process and the development of a position paper summarizing the NORM situation in Australia, a Safety Guide was developed, to enhance awareness and provide general guidance on NORM management. The Safety Guide recommends a graded approach to NORM management, based on exclusion, exemption, clearance and regulation. It includes sections on general radiation protection principles, impact assessment, assessment of the need for regulation, development of a NORM management plan, and annexes on NORM management in the oil and gas, bauxite processing and phosphate industries, written by experts from the industries. These industries were chosen because of their experience with NORM management and the availability of good data.

1. INTRODUCTION

Following the colonization of Australia by Europeans in various locations, the colonies combined in 1901 to become a Federation, which now comprises nine political jurisdictions. These are the Federal (Commonwealth of Australia) government, six State governments, and two Territory governments. Each jurisdiction has its own acts of parliament and regulations relating to radiation protection. These are different in detail, resulting in confusion with regard to radioactive waste management and control of radioactive sources. In addition, there are several Commonwealth agencies with an interest in NORM.

Mining has been an important part of Australia’s economy for more than 150 years. Most mineral ores, including coal, oil and gas, bauxite and mineral sands, contain radionuclides from the uranium and thorium and thorium radioactive decay chains in low, but variable, concentrations. Mining and processing of ores to extract minerals can alter the concentration of these naturally occurring radionuclides in products, by-products, residues and wastes.

This symposium has five themes:

- Theme 1: Operational radiation protection in workplaces with a potential for increased exposure;
- Theme 2: The extent to which general occupational, health and safety (OHS) and environmental regulation can provide a baseline level of protection of workers and the public against NORM exposure;
- Theme 3: Occupational radiation protection in non-production situations;
- Theme 4: Transport of NORM, including cross-border issues;
- Theme 5: Use and management of NORM residues.

This paper describes the history of NORM management in Australia, in the context of these five themes.

2. SOURCES OF NORM IN AUSTRALIA

The following paragraphs briefly describe the most common sources of NORM found in Australia and their uses, and associated management issues.

2.1. Extraction and/or processing of mineral ores

The mining industry produces very large volumes of NORM, particularly as residues and by-products. There is a strong economic incentive to use these large volumes of material, to avoid the costs associated with long term storage or disposal.

2.1.1. Uranium mining

Radium ore was first mined in Australia in 1906 at Radium Hill in South Australia. Uranium mining commenced at Rum Jungle, in the Northern Territory, in 1949. In the 1950s several small (underground and open pit) uranium mines were operated at Nabarlek and in the South Alligator River Valley, also in the Northern territory. These early mines were not regulated and the sites were not rehabilitated when mining ceased. Nabarlek has now been largely rehabilitated, while Rum Jungle and Radium Hill have undergone some remediation, and there has been a systematic programme of remediation of the abandoned sites in the South Alligator River [1].

The Ranger open pit mine (Northern Territory) opened in 1980. Mining of a very large copper deposit at Olympic Dam (South Australia) commenced in 1988. The copper was found to contain commercially viable levels of uranium, and Olympic Dam is now one of the world’s largest producers of uranium. An in situ leach uranium mine has opened recently at Beverley, (South Australia). These mines have been regulated since operations commenced.

2.1.2. Mineral sand mining

Australia is one of the world’s largest producers of heavy-mineral sands (rutile, ilmenite, and zircon). Mineral sand mines have been operating since the 1930s in New South Wales, Queensland and Western Australia, and more recently in Victoria. Large scale mineral sand mining commenced in the 1960s. Radionuclide concentrations in mineral sands vary considerably between mines. Most of the radionuclides encountered in mineral sands tend to be attached to the heavy minerals and therefore stay with the products. The most common method of mineral sand separation in Australia is the dry separation process, which does not cause changes in the solubility or mobility of individual radionuclides. Tailings from the dry separation process are therefore in the same physical form as the original ore and are returned to the original mining void. Tailings from the mineral sands separation process were used in the past as landfill. Regulatory processes now control the disposal of these materials.

Australian monazite is not traded because of economic considerations. In most cases the monazite concentrate is returned to the previously mineralized zone within the mining void.

2.1.3. Oil and gas production

Much of Australia’s crude oil and gas is produced by offshore drilling and extraction platforms in Bass Strait, the Northwest Shelf and the Timor Sea. These operations produce scales and sludges containing elevated concentrations of radium isotopes, which vary considerably from one production field to another. Although the quantities of NORM waste resulting from oil and gas production are small, these wastes are often contaminated with oil, and their safe management and disposal pose special problems. The cleaning and/or disposal of contaminated equipment can also be a significant problem.

2.1.4. Processing of phosphate rock: Fertilizer production and phosphogypsum

Australia uses very large quantities of fertilizer, mostly produced from imported phosphate rock. This leads to the production of large volumes phosphogypsum (by-product or waste) containing elevated levels of radium. Phosphogypsum is not currently used in Australia because of the ready availability of natural gypsum.

2.1.5. Bauxite processing and aluminium production

Australia produces approximately 40–50% of the world’s bauxite, mostly using strip mining or open cut methods. The radionuclide content of the bauxite ore is highly variable. NORM mud waste from bauxite mining (‘red mud’) is produced in large volumes. Disposal of this mud waste requires careful management as it is caustic and can present a significant environmental hazard.

2.1.6. Other minerals

The extraction and processing of other minerals such as copper, gold, iron ore, nickel, has expanded greatly from about 1950 onwards. Many of these ores and minerals are either themselves radioactive or contain radioactive contaminants and require appropriate management. These operations have produced large volumes of NORM residues and have resulted in a large number of legacy sites.

2.2. Coal mining and electricity generation (thermal power stations)

Australia generates most of its electricity by burning bituminous, sub-bituminous and brown coal, or lignite. The NORM wastes from coal burning (flyash and bottom ash) have traditionally been used as landfill and in road construction.

2.3. Water treatment

The use of groundwater for major public supply systems in Australia is not widespread. However, drinking water is treated in all Australian capital cities except Hobart, as well as regional centres and some small communities, to remove dissolved salts, heavy metals, salinity and soluble major elements such as calcium and magnesium.

The treatments used in Australia for surface and groundwater supplies remove radium and dissolved uranium contaminants quite efficiently. The main residues remaining from water treatment are flocculation sediments, filter sludges, other sand and sludges, spent ion exchange resins and reverse osmosis cartridges. The sediments and sludges are dried and disposed as landfill or by landspreading. Studies of radionuclide concentrations in Australian drinking water supplies indicate that drinking water treatment will not generate significant levels of NORM contamination in solid waste from the treatment plants.

2.4. Zinc smelter slag

Zinc smelter slag was used for some time as an abrasive medium for sandblasting in Queensland. However, the State regulator banned the use of the material in this way because of the high level of activity found in the raw material. As a result the producers were obliged to manage this material as a radioactive waste.

3. SCIENTIFIC STUDIES IN AUSTRALIA

In the early 1980s, studies of the speciation of radionuclides in soils and sediments were conducted [2], [3], and field studies were conducted around a titanium dioxide plant in Western Australia [4]. In the 1990s, studies on the use of red mud (from bauxite processing) as a soil conditioner were carried out [5]. The risks associated with the use of crushed granite as a termite barrier were also assessed [6]. A study of the potential risk associated with the use of phosphogypsum as a substitute for natural gypsum in plasterboard in Australian homes [7], [8] showed that the resulting increase in indoor gamma radiation exposures or indoor radon concentrations would not be significant. Similar conclusions were reached from a study of the radiological risk associated with the use of zircon glaze on ceramic tiles [9].

A series of studies was carried out in the Western Australia mineral sand mining and processing industry during the 1980s and 1990s [10]. Studies were also carried out at operational uranium mines [19], [20], and at non-operational sites [21]–[23]. Options for disposal of the wastes arising from oil and gas extraction and processing were also assessed [24].

4. REGULATORY DEVELOPMENTS UP TO 2002

During the period 1980–1995, a major review of the Western Australian mineral sands industry was carried out and regulations were introduced [25], particularly with respect to dust control. These changes led to substantial reductions in occupational doses.

By 2000, general (national) recommendations for limiting exposures to ionizing radiation had been introduced [26], and national codes of practice for the transport of radioactive materials [27], disposal of radioactive waste by the user [27] and near surface disposal of radioactive waste [27]–[29] had also been introduced.

The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) was established by a Commonwealth Act of Parliament in December 1998. ARPANSA, as a Commonwealth Agency, regulates Commonwealth entities and contractors, but has no jurisdiction within the States and Territories. ARPANSA’s tasks include promoting uniformity in the management of ionizing radiation in Australia. A National Directory for Radiation Protection [30] has been developed jointly by ARPANSA and the States and Territories, together with an evolving series of Standards, Codes of Practice, Safety Guides, and Recommendations dealing with specific topics in radiation protection. ARPANSA has systematically revised and updated many existing Codes [31], [32] and developed new documents [33] which are relevant to specific aspects of NORM management.

Two Committees provide advice to the CEO of ARPANSA. The Radiation Health and Safety Advisory Council (RHSAC) includes representatives from industry, universities, medicine, the public, and the CEO of ARPANSA, and provides advice on emerging issues and issues of major public concern in both radiation protection and nuclear safety. The Radiation Health Committee (RHC) includes all State and Territory regulators and the CEO of ARPANSA, and develops draft policies, Standards, Codes and Guidelines on radiation protection issues for the promotion of a uniform national approach. It is required to consult publicly in undertaking this role. The CEO of ARPANSA is also obliged to consider international best practice and to consult widely with technical experts, industry and the public when making regulatory decisions.

5. THE SITUATION IN 2002

By 2002, uranium and mineral sand mining and processing were regulated by the States and Territories, and the oil and gas extraction and processing, bauxite extraction and processing, and phosphate processing industries had well established operational and environmental radiation protection procedures in place. Other industries, including coal mining and electricity generation, mineral extraction and processing, water treatment, metal recycling, fertilizer manufacturing, and building materials did not have all these procedures in place. In general, the level of awareness of NORM issues in these industries appeared to be low. There were still issues relating to the differences between the Acts of Parliament and Regulations in the different jurisdictions.

6. DEVELOPMENTS SINCE 2002

6.1. Development of a NORM management strategy

In 2003, the CEO of ARPANSA formally requested the Radiation Health and Safety Advisory Council to provide advice on NORM. A nationwide consultation process was initiated in 2004, with the aim of preparing an advisory document containing a series of recommendations on the management of NORM in Australia. The consultation process included the preparation of a discussion paper [34], requests for submissions from industry, Government and the public, and the inclusion of a session on NORM at a National Conference on Radiation Protection and Radioactive Waste Management in Mining and Mineral Processing, held in Melbourne in 2005.

6.1.1. Outcomes of consultation

There was considerable industry support for national guidance, but there was concern in some industries that the aim of the process was to introduce an extra layer of regulation and a new regulator (in view of the existing level of environmental regulation). There was a need for better data in some industries and a systematic risk analysis before consideration of regulatory options. There was concern that negative economic impacts should be avoided, and that labelling materials as ‘low level radioactive material’ could have a significant negative impact in some industries. The idea that the criteria for regulation should not be based only on activity concentration, but should include consideration of risk, was strongly supported. There was support for awareness raising, but in consultation with stakeholders. Considerable extra data were provided in some submissions.

The Council's advice [35], which was presented to the CEO in September 2005 together with a report [36] summarizing NORM in Australia (industries, types of material, quantities of material, etc.), incorporated all this information.

The major recommendations of the Council were that:

(a) ARPANSA should develop national guidance on NORM management, in consultation with industry, and the States and Territories (through the Radiation Health Committee). This should include uniform exclusion and exemption provisions, guidance on the treatment and disposal of NORM residues and wastes, and guidance on remediation of contaminated sites;
(b) International guidance should be taken into account, for instance IAEA Safety Standards No. RS-G-1.7 [37];
(c) Additional data should be considered and sought if necessary;
(d) Industries requiring active NORM management should be identified and guidance for those industries developed;
(e) Where necessary, further requirements for the National Directory for Radiation Protection should be developed (after regulatory impact assessment);
(f) ARPANSA should develop a strategy to raise public awareness of NORM, and awareness of NORM management in relevant industries; this process should include consultation with industry and State and Territory regulatory authorities.

The CEO agreed with these recommendations [38], and directed that a Safety Guide on NORM management be prepared, and that:

(1) Guidance and regulation must be based on real industry data, a risk assessment and a graded approach;
(2) A stakeholder consultative group (including representatives from both Government and industry) be set up to help guide the project;
(3) Awareness raising should be carried out initially via a central web site, and in consultation with the stakeholder group and representatives from the States and Territories.

6.2. Development of the Safety Guide

A working group, comprising three scientists (two with considerable regulatory experience), and experts from the aluminium, oil and gas, and plasterboard (phosphate) industries, began work on the Safety Guide in March

2007. A draft document was considered by the Radiation Health Committee in March 2008, and released for public comment in May–June 2008. The final draft was approved by the RHC in July 2008, and the RHSAC recommended adoption of the Safety Guide in August 2008 [39].

6.3. RPS 15 — Safety Guide for the management of NORM

The Safety Guide contains an Introduction, and sections on industries where NORM radiation protection issues may arise, radiological issues in NORM management, regulatory issues in NORM management, operational issues (the NORM Management Plan, or NMP), remediation of legacy sites and a summary. It also contains references, an extensive bibliography, a glossary, and annexes on NORM management in the oil and gas, bauxite/aluminium, and phosphate industries.

In general, the approach to NORM management outlined in the Safety Guide follows that recommended by the IAEA. The term TENORM is not used, and the term NORM covers all materials containing naturally occurring radionuclides, and for which the original radionuclide concentrations and/or exposures have been modified by human activities.

The Safety Guide also recommends a graded approach to the regulation of NORM, based on the assessed level of risk to the environment, workers and members of the public, and the concepts of exclusion, exemption and clearance.

6.4. Industries where NORM radiation protection issues may arise

The uranium mining and processing and mineral sand mining industries were excluded from the scope of this Safety Guide as they were already regulated. Other industries where NORM may be a potential radiological issue were those discussed earlier.

The oil and gas, bauxite/aluminium, and phosphate industries are covered in the existing Annexes to the main document. These industries were selected because they have comprehensive data sets available and have considerable experience in radiation management.

6.4.1. Radiological issues in NORM management

This section includes guidance on exposure pathways, mineral extraction, mineral and downstream processing, transport of bulk commodities, residues and wastes, use of products containing NORM, management of NORM residues and wastes, and public perception.

6.4.2. Regulatory issues in NORM management

This section includes guidance on international developments in NORM management, current regulatory frameworks in Australia, assessing the need to regulate NORM, the graded approach to regulation, an iterative approach to dose and impact assessment, management of NORM wastes and residues, transport, site remediation and close-out requirements, and assessing the impact of regulatory proposals.

6.4.3. Assessment of the need for regulation of NORM and the graded approach to regulation

This section, which is probably the most important part of the Safety Guide, provides guidance on a sequence of steps.

NORM operations can be identified by the operator or the regulator. The operation should then be analysed to identify potential issues. Using a methodology established by discussion between the operator and the regulatory authority, the operator should then carry out a screening assessment. This assessment should be subjected to careful review.

If the screening assessment indicates that the radiological risks are negligible and are likely to remain negligible throughout the lifetime of the operation and during and following closure and any remediation required by the regulatory authority, the operator may be granted unconditional exemption from regulatory control. If the screening assessment indicates that unconditional exemption may not be allowed, the operator should carry out a more detailed assessment, again in consultation with the regulator.

In some cases the detailed assessment may indicate that unconditional exemption is appropriate, or that the risks are significant enough to require the operator to obtain a licence and operate under full regulatory control. At present, one operation in Australia is regulated in this way. For the remaining cases, the regulatory authority may choose to grant a conditional exemption. This could include provision for regular monitoring and reassessment to confirm that the basis for exemption remains valid.

Operations which are currently under regulatory control are required to have an approved Radiation Management Plan (RMP) in place. For those NORM operations that are carried out under conditional exemption, the Safety Guide recommends that the operator and regulator should negotiate an operation-specific NMP, which should be similar to the RMP used in those operations that are under full regulatory control.

This basic NORM management process is summarized in Fig. 1.

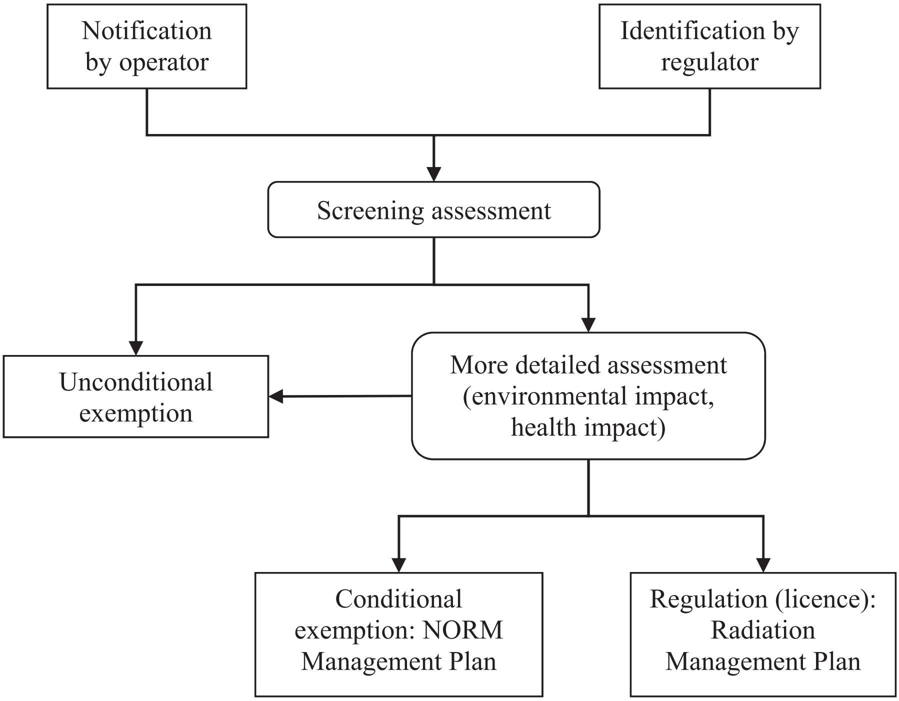

FIG. 1. The basic NORM management process.

6.5. Operational issues — the NORM Management Plan

The development and implementation of the NMP should help to promote awareness and encourage a sound safety culture. The key elements of the NMP are identification of potential sources of health impact on workers, members of the public and the environment, management of the health impact on workers, members of the public and the environment, remediation and close-out requirements for operational sites, and non-radiological issues such as chemicals and heavy metals. The NMP should be developed by negotiation between the operator and the appropriate regulatory authority.

6.6. Remediation of legacy sites

The major issues associated with these sites include the following:

(a) Often little or no documentation of activities at the site is available;
(b) It is difficult to assign responsibility for any cleanup that may be required;

(c) Characterization of the site can be difficult — in many cases the demographics have changed, and sites that were used for industrial activity are now used for residential purposes;
(d) Impact assessment can be complicated, for the same reasons that make site characterization difficult.

The Safety Guide recommends the development and implementation of an iterative approach to impact assessment and remediation of legacy sites. This approach starts with a screening assessment; the subsequent strategy depends on the outcome of this initial assessment. If the screening assessment indicates that the environmental and health impacts of the site are negligible, no further action is required. Otherwise, a remediation plan is developed and its predicted impact is estimated. If necessary, the plan is modified until the predicted impact is acceptable. The plan is then implemented, and monitoring is carried out to check for deviations from the initial predictions. If there are significant deviations the process should be modified to bring the predicted and measured impacts back to acceptable levels.

This process is designed to build confidence among all stakeholders in the ultimate outcome. It is critically important to involve all stakeholders at all stages of this process.

6.7. Annexes

The Safety Guide currently contains three annexes, which discuss management of NORM in the oil and gas, bauxite/aluminium and phosphate industries. Each annex gives a brief description of the industry, and a description of the raw materials, processing steps, management of the NORM product and waste/residue streams, operational radiation protection issues, and relevant downstream processing, environmental and public health issues.

7. FUTURE CHALLENGES

The current plan is to provide additional annexes dealing with coal extraction and electricity generation, and metal extraction. Further annexes will be added as appropriate. These could include an annex on safety assessment and an annex on environmental impact assessment. In addition, development of the overall NORM management framework will continue, and an ARPANSA NORM web page will be developed (in consultation with the Stakeholder Group) as a resource for information on NORM.

8. SUMMARY

The graded approach to regulation is extremely important when dealing with NORM. In some cases (uranium and mineral sand mining and processing) there is a clear and demonstrable radiological impact. In other cases, raising awareness without causing unnecessary alarm is extremely important. It is important to emphasize that the aim is to enhance safety where there is a clear need to do so.

General OHS procedures can provide a measure of protection against NORM exposure. Dust suppression and the use of masks or respirators in potentially dusty environments can significantly reduce dust inhalation. Normal occupational hygiene practices can also be effective in limiting exposures in workplaces where NORM is present. For external exposure, inhalation of radon and ingestion of radionuclides in food, general OHS procedures are less effective. Management of NORM products, by-products, wastes and residues is not always easily amenable to general OHS and regulatory procedures because of the widespread use of these materials and the very long lifetimes of the radionuclides.

General environmental regulation can also contribute to protection of workers and the public by limiting discharges from processing facilities, and inhibiting the mobility of waste materials. However, such regulation may not always be effective, because the radionuclides in NORM are long-lived, and the relative importance of different exposure pathways can change with time. This type of regulation does not usually deal with the problem of responsibility for remediation of legacy sites.

Occupational radiation protection in non-production situations can apply to office staff in uranium mines, drivers of transport vehicles, workers in the waste disposal and building industries, workers involved in remediation of legacy sites, and people involved in trading products, by-products and commodities containing NORM. The recommendations in the NORM Safety Guide cover the handling of NORM in these situations.

Australia’s federal structure means that cross-border issues can arise for transport between states as well as between countries. The updated Transport Code [31] and accompanying Safety Guide [33] address these issues.

In Australia, red mud is used as a soil conditioner and in some landfill applications, and fly ash is used in building materials and in landfill. Phosphogypsum is not currently used in building materials, and therefore has to be stored. The oil and gas industry has an ongoing problem in managing sludges and scales, from both occupational and environmental standpoints, in terms of storage/disposal/cleaning of contaminated pipes and equipment, and disposal of contaminated scales and sludges.

Australia has a large number of NORM legacy sites that may require assessment and possibly remediation. Some of these sites are also contaminated with chemicals and heavy metals. Many of the old uranium mine sites, particularly those in the Northern Territory, have been or are being remediated [1].

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PRINCIPLES FOR SPECIFIC CLEARANCE USED IN PROPOSED NORM REGULATIONS

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Abstract

The Swedish Radiation Safety Authority has launched a project in order to establish regulation for exemption and clearance. Regulations are planned soon to be promulgated for exemption and clearance of NORM waste. The main sources by volume of NORM in Sweden are products related to alum shale: (i) the shale itself, as it may be present in building activities and well drilling, (ii) old remnant heaps of shale residue from oil extraction and mixtures of shale and limestone, and (iii) alum shale based concrete from dismantling old buildings. In this work, the Swedish Radiation Safety Authority is in accordance with the expected new European Union Basic Safety Standards, which will include provisions for NORM. The concept of intrusion into a disposal site where NORM waste has been disposed as cleared or exempted waste is not in accordance with the recommendations given in Publication 81 of the International Commission on Radiological Protection regarding the dose criterion of 10 mSv/a below which no action would be warranted in case of intrusion. This contradiction also prevails for other low level near-surface sites in Sweden. However, the main waste streams in Sweden involve material with relatively low concentrations of radionuclides, mainly uranium, or with small volumes, so that the dose calculations are relatively insensitive to the choice of scenario.

1. INTRODUCTION

In Sweden, a project is under way to define exemption levels for waste comprising naturally occurring radioactive material (NORM), including regulations for specific clearance in connection with disposal on municipal disposal sites and for restricted forms of recycling. Specific clearance, also called conditional clearance, is clearance of a particular waste stream and could therefore also be called ‘(specific) waste stream clearance’. The focus here is on the disposal of NORM in a municipal waste disposal site or any other surface site with similar (post-closure) protective capability, or on a restricted form of recycling.

2. NORM INDUSTRIES IN SWEDEN

Sweden has few industries where NORM is produced as a by-product. The only industry where raw material containing naturally occurring radioactive substances is processed to create a product is the steel industry. The concentration of radioactive substances in the raw material for the steel industry is low, however, and the concentration of ^{238}U in sludge is up to 0.25 Bq/g. Industries using large amounts of water may have problems with deposits of naturally occurring radionuclides in pipes. Deposits are usually found when pipes are transported to scrap metal yards for recycling. Also, ash from peat combustion will enrich radionuclides to concentrations which require the material to be handled as NORM. A growing issue is the volume of water treatment filters enriched with radionuclides as there are recommendations on uranium in drinking water. The amount of NORM is in general quite small, but occasionally the concentration can be high. The exception is peat ash, the amounts of which are much larger than other enriched material and which also contains ^{137}Cs.

Historical waste from industry processes is found in several places. Large amounts of burned alum shale are found in piles in former alum shale mining areas. The red coloured material is reused as a filling material for tennis courts and sports grounds. Residues from the mining and steel industry can also be found, as well as phosphogypsum from the phosphate industry. There is also a large amount of lightweight concrete based on alum shale in existing buildings. When the buildings are demolished one day, the handling of the material must be regulated as the radionuclide concentration is above the relevant exemption level. There is a large amount of historical material and most of it is stocked in piles, which are now covered with vegetation. The ^{238}U activity concentrations can be up to 2.5 Bq/g, but the activity concentration is usually more moderate [1].

3. EXEMPTION IN CONNECTION WITH NORM

The knowledge on existing NORM industries and enriched material in Sweden forms a basis for regulations on NORM management. The proposed regulations are based on a graded approach as described in Fig. 1 and suggested by the European Commission [2]. According to the Radiation Protection Act and Ordinance, the radiation safety authority has the mandate to issue more specific binding regulations, as well as general advice. It is foreseen that perhaps not all the options in Fig. 1 will be used in the regulations.

Although the current Radiation Protection Act includes NORM, the industries are not licensed at the moment, even though the activities exceed the exemption levels. Licensing of activities involving NORM is not considered to be

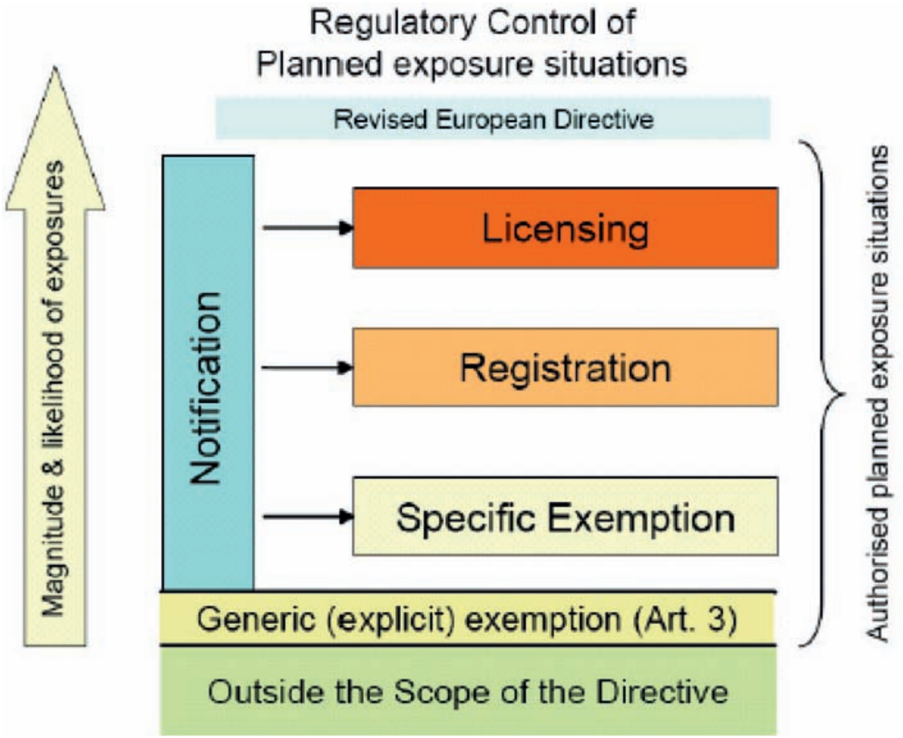

FIG. 1. Graded approach to regulation.

necessary at the same level as traditional practices, and it is proposed that the criterion for licensing will be ten times higher than currently specified — for the ^{238}U decay series, the new criterion would be 10 Bq/g.

It is proposed that the general approach to regulation should be to use an activity ‘ladder’ for specific clearance of material (see Fig. 2). The proposed regulations do not include radiological protection of workers. The general clearance levels for NORM will be, as proposed in the new European Union basic safety standards, the same as the exemption levels. Nevertheless, one has to bear in mind that there might be other restrictions on the use of the material — in the case of building material, for instance, there are building codes that impose controls on radon concentration and gamma exposure indoors. Historical materials, material from bed rock drilling and building materials with activity concentrations of up to three times the exemption levels can be reused in applications such as landfill and road construction. The activity concentrations in historical materials are known. For drilling material, the amounts are small and there is a natural mixing of materials with lower activites.

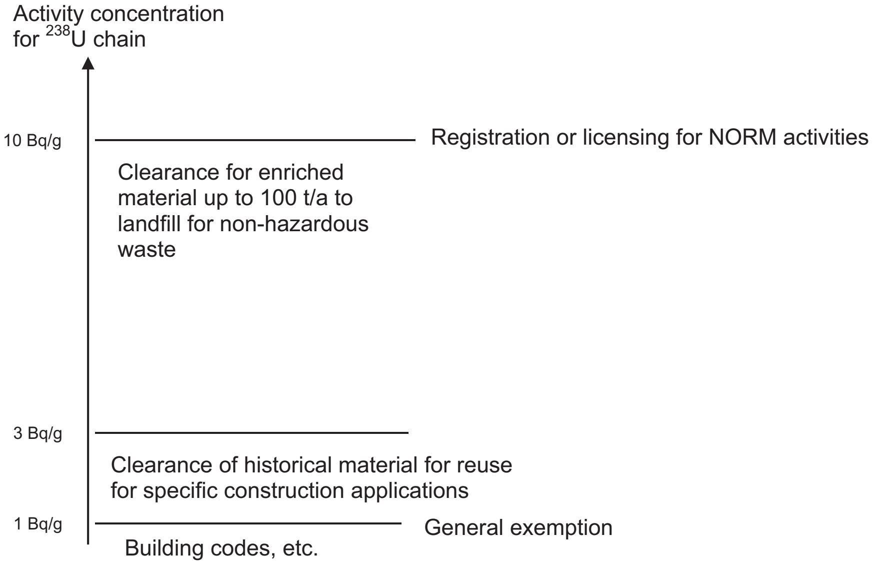

FIG. 2. Specific clearance for NORM.

Materials which have become enriched in radionuclides, such as water filter residues, scale and peat ash, may be deposited at landfill facilities constructed for non-hazardous waste in quantities of up to 100 t/a. The activity concentration limit for such disposal is proposed to be 10 Bq/g for the ^{238}U chain, which corresponds to the limit for uranium in environmental regulations concerning chemical toxicity. The proposed regulation exempts the material from further control and no environmental monitoring will be required.

4. INTRUSION

The process of setting criteria for specific clearance in connection with the disposal of radioactive material presents a problem, particularly for long lived radioactive waste. In the process of setting maximum permitted levels, for instance, for disposal in a municipal disposal site, the regulatory body must make assumptions regarding future exposure scenarios such as those related to future (inadvertent) intrusion.

The recommendations of the International Commission on Radiological Protection (ICRP) in its Publication 81 [3] are of limited value for this type of waste management. NORM disposed of in a municipal disposal site is both long lived and solid radioactive waste, so the activity should be covered by the scope of the recommendations. The recommendation for the dose rate for the intrusion scenario is described by the ICRP as follows:

> "The Commission considers that in circumstances where human intrusion could lead to doses to those living around the site sufficiently high that intervention on current criteria would almost always be justified, reasonable efforts should be made at the repository development stage to reduce the likelihood of human intrusion or to limit its consequences. In this respect, the Commission has previously advised that an existing1 annual dose of around 10 mSv per year may be used as a generic reference level below which intervention is not likely to be justifiable. Conversely, an existing annual dose of around 100 mSv per year may be used as a generic reference level above which intervention should be considered almost always justifiable."

A precise interpretation of ICRP Publication 81 in this situation would lead one to conclude that intrusion should lead to an annual dose greater than 100 mSv in order to always justify action to reduce the consequences. The ICRP recommends a dose range of 10–100 mSv/a for regulatory consideration of such actions. Clearly these dose rates are too high for a municipal waste disposal site. Perhaps higher doses could be accepted as a hypothetical exposure from radioactive waste in a geological disposal site. However, one must acknowledge that there are question marks when specific clearance is equated with radioactive waste disposal, although in some cases disposal on municipal disposal sites is accompanied by requirements that radioactive effluents are measured. Such a requirement is not in harmony with the concept of clearance or exemption, which assumes that no regulatory action needs to be taken to control cleared or exempted NORM material. The conclusion is therefore that disposal sites for cleared or exempted waste do not fit well into the description of waste disposal. To some extent, the philosophy of some near-surface low level disposal sites may also have to be modified, regarding treatment of human intrusion.

In the regulations currently proposed, the above-mentioned problem does not present a serious obstacle since the levels suggested are below 800 ppm for natural uranium, the main radioactive element of concern in NORM waste in Sweden. Calculations show that, for a relatively wide choice of scenarios, the dose would still not exceed a regulatory criterion of 0.3 mSv/a.

5. RADIOACTIVE WASTE ARCHIVES

Active and passive controls, including systems for long term conservation of information, have an obvious place when radionuclides with half-lives of 30 years or less are the dominant radionuclides in radioactive waste. The picture is less clear for NORM waste with long lived radionuclides such as ^{238}U.

6. LONG TERM RECORD KEEPING IS PART OF OPTIMIZATION OF PROTECTION

For long lived NORM waste, that is, waste containing long lived radionuclides of natural origin, the value of record keeping is not as obvious. However, the impossibility of guarantees for long term enforcement of controls does not constitute a reason for not conserving information. On the contrary, optimization as understood by the ICRP in recent years requires that we should ask the question “have I done everything I reasonably can to limit doses?” Obviously one cannot give a positive answer to the question unless relatively simple measures are taken, such as the establishment of a radioactive waste archive.

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HUMAN EXPOSURE TO RADIOACTIVITY FROM MINING AND INDUSTRIAL RESIDUES[1]

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Abstract

Radionuclides of natural origin are present in all soils and rocks. UNSCEAR 2000 reports concentrations of natural radionuclides in a wide variety of materials and with a wide range of activity levels. Exploration and processing of these resources and production of consumer items can lead to further enhancement of the radioactivity in the products, by-products, residues or wastes, arising from industrial processing. A conceptual difficulty in the management of NORM has arisen, largely because there have been different perceptions of the potential radiological hazard since all of these materials are naturally radioactive but, in large part, are not associated with the nuclear industry. The IAEA, recognizing the practical problems of regulating large quantities of NORM materials with low levels of radioactivity, proposed that regulatory criteria for radionuclides of natural origin should be based on the upper end of the worldwide distribution of natural radionuclides (as described by UNSCEAR, for example). The IAEA Safety Guide RS-G-1.7 suggests that “doses to individuals as a consequence of these activity concentrations would be unlikely to exceed about 1 mSv in a year, excluding the contribution from the emanation of radon, which is dealt with separately in the BSS”. However, in order to evaluate this hypothesis, the IAEA decided to perform independent calculations of dose associated with a ‘generic’ hypothetical NORM residue deposit. This paper describes the methods, data and results of that study. The study develops the characteristics of a ‘generic’ NORM residue deposit of 2 million m^3 covering 10 ha that contains the natural uranium (^{238}U) decay series radionuclides and/or the natural thorium (^{232}Th) decay series radionuclides. The radionuclides are assumed to be in equilibrium, each at a concentration of 1 Bq/g. To the extent practicable, the doses to adults and children (1 year old child) living close to such deposits were assessed using the models and assumptions of IAEA Safety Reports Series No. 44. The possibility that some NORM residues may be acid generating was also considered. Based on the findings of this study, while doses slightly in excess of 1 mSv/a are possible, it is considered that the dose to an adult or child who lives in close proximity to a NORM residue deposit would be well below 1 mSv/a for the vast majority of real situations.

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[1] Work performed within the International Atomic Energy Agency’s programme on Developing Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources.

1. INTRODUCTION

1.1. Background

NORM is an acronym for naturally occurring radioactive material, which contains radionuclides from the so-called ‘primordial’ decay series resulting from the decay of the ^{238}U, ^{235}U and ^{232}Th decay series and from other long lived radionuclides such as ^{40}K. In the majority of situations, the naturally occurring radionuclide concentrations are not sufficiently elevated to pose a potential hazard to people. In some cases however, where the radionuclide concentrations are much higher than the normal range of background levels, there may be potential for correspondingly higher doses to people.

Although attempts have been made to distinguish between materials in their natural state and materials in which natural radioactivity has been enhanced by man, for present purposes, as is the IAEA’s practice, the term NORM is used in this report regardless of the state of the material.

UNSCEAR [1] reports concentrations of natural radionuclides in a wide variety of materials and with a wide range of activity levels. Examples of ores that have been found to be associated with anomalous (high) levels of NORM include ores of uranium, tin, tantalum, niobium, rare earths and aluminium, some copper and gold occurrences and phosphate rock. Mining and processing of these resources and production of consumer items can lead to further enhancement of the radioactivity in the products, by-products, residues or wastes, arising from the industrial processing. A few examples of radioactivity levels in a variety of NORM materials are summarized in Table 1. Concentrations of these radionuclides may be modified in the residues or by-products arising from the processing of the materials. Examples of this include radioactivity levels in phosphogypsum from the production of phosphate fertilizer, oil scale wastes and residues from the production of titanium dioxide among many others.

A conceptual difficulty in the management of NORM has arisen largely because there has been a perception of potential radiological hazards with these materials, common both in the industries concerned and in the public, since all of these materials are naturally radioactive.

For purposes of determining the need for regulatory control, the IAEA established radiological criteria for radionuclides of both artificial and natural origin in materials. These criteria are specified in IAEA Safety Standards Series No. RS-G-1.7 [3]. For radionuclides of natural origin, Ref. [3] concludes that the derivation of activity concentration values on the basis of the same radiological criteria as those used for artificial radionuclides is not practical and in many cases, would produce values lower than concentrations occurring in the natural environment. In view of this, it was proposed that radiological criteria for

TABLE 1. EXAMPLES OF RADIOACTIVITY LEVELS IN NORM
(adapted from Ref. [2])

	Radionuclide with highest activity concentration	Typical activity concentration (Bq/g)
Monazite sand	^{232}Th series	40–600
Metal ores, e.g. Nb/Ta, Cu, Au	^{238}U and ^{232}Th series	Up to 10
Zircon sand	^{238}U series	2–4
Phosphate rock	^{238}U series	0.03–3
TiO_2 feedstocks	^{232}Th	0.001–2
Bauxite	^{232}Th series	0.035–1.4
Red mud (alumina production)	^{238}U, ^{232}Th	0.1–3
Phosphogypsum (H_2SO_4 process)	^{226}Ra	0.015–3
Niobium extraction slag	^{232}Th	20–120
Tin melting slag	^{232}Th	0.07–15
Scale (oil and gas production)	^{226}Ra	0.1–15 000
Residue (rare earth extraction)	^{228}Ra	20–3000
Scale (TiO_2 pigment production	^{228}Ra, ^{226}Ra	<1–1600
Scale (rare earth extraction)	^{226}Ra, ^{228}Th	1000
Sludge (oil and gas production)	^{226}Ra	0.05–800
Residue (niobium extraction)	^{228}Ra	200–500
Coal	^{238}U and ^{232}Th series	0.01–0.025
Scale (coal mines with Ra rich inflow water)	^{226}Ra, ^{228}Ra	Up to 200

radionuclides of natural origin should be based on the *upper end* of the worldwide distribution of natural radionuclides (as described by UNSCEAR [1], for example). Reference [3] goes on to indicate that:

- “It is usually unnecessary to regulate…” material containing radionuclides of natural origin at activity concentrations below 1 Bq/g for radionuclides in the uranium and thorium decay series and below 10 Bq/g for 40K; and
- “Doses to individuals as a consequence of these activity concentrations would be unlikely to exceed about 1 mSv in a year, excluding the contribution from the emanation of radon, which is dealt with separately in the BSS”. (emphasis added)

1.2. Objective

As indicated above, doses received by people living near NORM residue deposits with activity concentrations of the natural uranium and thorium decay series of 1 Bq/g or less are considered to be below 1 mSv/a [3]. However, as indicated in Table 1, there is considerable variability among NORM residues. Thus, the IAEA decided to perform independent assessments of dose associated with such NORM residue deposits to confirm this position. This paper provides an overview of the results of that study [4].

1.3. Study approach

The key elements of the NORM residue study were to:

(a) Consider reasonably available information from real world examples of actual NORM residue deposits that could be used to benchmark representative NORM residue deposits;
(b) Define the characteristics of the representative NORM residue deposits to the extent possible, based on real world experience;
(c) Use the modelling approach and assumptions set out in Ref. [5], where appropriate;
(d) Develop exposure pathways and dose assessment for the representative NORM residue deposit following IAEA Safety Reports Series No. 44 [5] where appropriate;
(e) Discuss the potential doses arising to a person(s) living in close proximity to such a NORM residue deposit.

The key assumptions used in the study were:

(1) A nominal residue deposit volume of 2 million m^3 covering 10 ha;
(2) Radionuclides in the natural uranium (^{238}U) decay series or the natural thorium (^{232}Th) decay series, or both, are present in the residual materials at a concentration of 1 Bq/g;
(3) Within the residue deposit, all of the decay products within the ^{238}U and the ^{232}Th decay series are assumed to be in equilibrium with the parent radionuclide;
(4) The NORM residue deposit could potentially be acid generating.

2. CHARACTERISTICS OF THE REPRESENTATIVE NORM RESIDUE DEPOSIT

NORM residues are quite variable and depend in large part on the geological setting of the source material, the specific uranium and/or thorium content of the materials, and the effects of processing (concentration or mobilization of radionuclides in some cases).

For most unaltered waste materials, radionuclides are mobilized with weathering and the passage of water and air through the pile. Some of the radionuclides will transport more readily than others, which may be precipitated or sorbed on surfaces of waste materials and local subsurface soils. Retardation of the movement of radionuclides such as radium and uranium has been demonstrated at many locations and retention coefficients, although highly variable, have been reported for many soils and rock types.

Information regarding the characteristics of NORM residue deposits was obtained from IAEA publications, open literature and journal papers, proceedings of workshops and conferences, and in-house reports. Full details with citations are provided in Ref. [4]. The information was evaluated and used to develop the characteristics of a nominal (‘representative’) NORM residue deposit as described briefly below.

2.1. Physical characteristics

There is a large variety of NORM residues arising as the result of the processing of ores containing NORM, including tailings and treatment sludges; various slags from the production of tin, niobium and tantalum ores; as well as phosphogypsum, a by-product of fertilizer production. Moreover, the modelling of bulk amounts of material such as NORM residue deposits requires many assumptions to be made about the quantity of material stored or disposed of, the location, where it is placed relative to the public, and the characteristics of the environment which affect the pathway calculations, especially that of the water pathways. All these quantities are highly variable and site-specific. Thus, assumptions are needed to characterize a representative NORM residue deposit. For purposes of this assessment, the NORM residue deposit is assessed to have the characteristics shown in Table 2.

2.2. Radioactivity

Activity concentrations vary, depending on the type of mining activity, industrial processing and the type of NORM residue. Some non-uranium mining residues have elevated uranium activity concentrations. As noted previously in

TABLE 2. PHYSICAL CHARACTERISTICS OF REPRESENTATIVE NORM RESIDUE DEPOSIT

Parameter	Value
Assumed deposit area	100 000 m^2
Assumed deposit volume	2×10^6 m^3
Assumed distance to residential area	20 m
Assumed thickness of contaminated zone	20 m
Calculated total mass of contaminated material	3.6×10^{12} g

Ref. [3], “It is usually unnecessary to regulate…” material containing radionuclides of natural origin at activity concentrations below 1 Bq/g for radionuclides in the uranium and thorium decay series. Thus the present analysis assumes that exposure is due to natural uranium (U_{nat})[2], ^{230}Th, ^{226}Ra, ^{210}Pb, ^{210}Po, ^{232}Th, ^{228}Th and ^{228}Ra. The activity concentrations of each radionuclide in the residue deposit were taken to be 1 Bq/g.

2.3. Leachate

Overall, the factors controlling the solubility of key radionuclides in NORM residues are reasonably well known but the actual characteristics of leachate vary greatly. Laboratory and field data from various sites show that the rates of contaminant release from waste rock change over time. In cases where there is no sharp change in pH, that is, where the system remains neutral, contaminant release rates generally decrease over the long term [6].

Based on the information reported in Ref. [4], the data in Table 3 were suggested as providing a reasonable characterization of the radiological characteristics of leachate from representative NORM residue deposits. Data are provided for both non-acid-generating and acid generating residues. Although some of the assumed leachate characteristics differ by a factor of up to about 10, this distinction is typically expected to reduce quite rapidly in most situations as alkalinity in the receiving environment neutralizes the leachate as it moves through soils and rocks. Although data are limited, it is thought that these characteristics are in the range of the upper 10th percentile values.

For the radioactivity levels given in Table 3, a set of realistic K_d values was estimated and is provided in Table 4. These values are considered to reasonably

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[2] U_{nat} denotes ^{238}U with ^{234}U in equilibrium and ^{235}U at its natural abundance ratio.

TABLE 3. REPRESENTATIVE SOURCE CONCENTRATIONS FOR NORM RESIDUE DEPOSITS

	Typical NORM residue, non-acidic	Typical NORM residue, acidic[a]
U and Th content:		
U_3O_8	0.01%	0.01%
^{232}Th	0.01% (0.46 Bq/g)	0.01% (0.46 Bq/g)
Leachate quality:		
^{238}U	1 mg/L (12 Bq/L)	10 mg/L (124 Bq/L)
^{226}Ra	1 Bq/L	1 Bq/L
Th	<0.01 mg/L (<0.041 Bq/L)	0.1 mg/L (0.41 Bq/L)
^{210}Pb [b]	0.2 Bq/L	1 Bq/L
^{210}Po [b]	0.05 Bq/L	0.1 Bq/L

[a] It is highly unlikely that a person would have an acidic water supply. Constituents such as pH, salinity, iron levels and metals would result in aesthetic characteristics which are likely to preclude the use of the water.

[b] Based upon seepage/groundwater monitoring data for a uranium mine.

TABLE 4. ESTIMATED K_d VALUES FOR A REPRESENTATIVE NORM RESIDUE DEPOSIT

	K_d (mL/g)	
	Realistic modelling	US DOE RESRAD modelling for sandy material
Lead	5000	550
Polonium	2000	3000
Radium	1250	9100
Thorium	1000	5800
Uranium	50	1600

describe K_d values of a representative NORM residue deposit. However, given that K_d values vary considerably in literature, it was considered informative to consider alternative values to examine how sensitive the doses are to the selection of the K_d. A great deal of information, including information on K_d (see for example, Ref. [7]), has been compiled in support of the US DOE RESRAD modelling system for estimating doses. The K_d values from RESRAD data for sandy material provides the lower bound for all the data cited, and as such, tends to give a very conservative (high) estimate of groundwater concentration. Thus, for present purposes, the assessment of the NORM residue heaps was also performed using the sand K_d values from the RESRAD data collection.

2.4. Transport in groundwater

The transport of radionuclides in groundwater has been studied extensively. Almost without exception, rocks and soils attenuate radionuclides where concentrations in the source exceed background levels.

The transport process of soluble materials in water generally has two components, advection and dispersion. Advection is transport along with the average pore water velocity. Dispersion is transport within the water, due both to molecular diffusion and small scale differences in flow speeds. When the pore water velocity is fast, the dispersion component is not a significant contributor to the movement of the soluble chemicals. This is particularly true when the groundwater is moving in coarse sandy soil, where the pore water movement is relatively fast, and the retardation factor is relatively small. A range of hydraulic conductivities and permeabilities for various rocks and unconsolidated deposits has been published [8]. The hydraulic conductivity K ranged from 0.1 m/s for coarse gravel to 10^{-13} m/s for metamorphic and igneous rock. It is expected that the hydraulic conductivity for residue materials with large particle sizes would have a wide range, depending on profiles and surface conditions of the residue deposits, the nature and size range and size segregation of the materials, pore volumes, compaction as well as climatic conditions such as freeze–thawing, wetting and drying.

As will be seen later, the dose from NORM residue deposits is strongly dependent on the seepage from the deposits and consequent groundwater concentrations. The dose calculations for these groundwater pathways were based on Scenario RW in IAEA Safety Reports Series No. 44 [5]. This model assumes that radionuclides within the residue deposits are available for migration into the aquifer. The rate at which the radionuclides move is determined by a distribution coefficient model, which takes into account leach rates and uses a retardation factor to determine the seepage and water. As a ‘test of reasonableness’, radionuclide concentrations in seepage, estimated using the equations from Ref. [5], were compared with the previously-mentioned nominal seepage concentrations as shown below in Table 5.

TABLE 5. MEASURED AND ESTIMATED RADIONUCLIDE CONCENTRATIONS IN SEEPAGE

	Nominal concentration (Bq/L)		Estimated concentration (Bq/L)	
	Non-acidic	Acidic[a]	Realistic distribution coefficients	Sandy distribution coefficients
^{238}U	12	124	20	28
^{230}Th	—	—	1.0	0.31
^{226}Ra	1	1	0.80	2.0
^{210}Pb	0.2	1	0.20	3.7
^{210}Po	0.05	0.1	0.50	6.7
^{228}Th	—	—	1.0	0.31
^{232}Th	<0.041	0.41	1.0	0.31
^{228}Ra	—	—	0.80	2.0

[a] It is highly unlikely that a person would have an acidic water supply. Constituents such as pH, salinity, iron levels and metals would result in aesthetic characteristics which are likely to preclude the use of the water.

3. DOSE ASSESSMENT

This section deals with exposure to radiation from a representative NORM residue deposit. For the purposes of this assessment, the modelling approach described in Ref. [5] was used as the basis of exposure estimates where appropriate and the realistic case parameters are used unless otherwise indicated. It should be noted that, for the numerical criteria presented in IAEA Safety Guide RS-G-1.7 [3], radon is specifically excluded from the calculations. However, separate calculations were performed to provide an assessment of the likely radon concentrations at a residence near a NORM residue deposit for comparison with the reference level for radon recommended in the 2007 Recommendations of the ICRP [9]. The results, reported in Ref. [4], show that the radon concentration was well below the reference level.

For this assessment, people were assumed to live very close to the NORM residue deposit and, for consistency with the modelling approach used in Ref. [5], an adult and a one year old child were chosen as (hypothetical) receptors. The exposure pathways considered in this assessment include:

(a) Inhalation of dust;
(b) External exposure (from dust depositing on the ground at the residence);

(c) Ingestion of dust (from dust depositing on the ground at the residence);
(d) Ingestion of garden and agricultural products (from irrigation with contaminated groundwater);
(e) Groundwater pathway;
(f) Surface water pathway.

The pathway calculations were developed from those provided in Ref. [5], which details the evaluations necessary when dealing with radioactivity from bulk amounts of solid materials for which regulatory control may or may not be warranted. The modelling approach described in Ref. [5] uses generalized scenarios and assumptions only for radionuclides of artificial origin and not for radionuclides of natural origin. This study extends this scenario-based approach to radionuclides of natural origin. For the purposes of this assessment involving exposure scenarios for the representative NORM residue deposit(s), the modelling approach and realistic case assumptions from Ref. [5] were used where appropriate, unless otherwise indicated.

Overall, the key parameters related to the physical characteristics of the waste heap and residential area were chosen according to the realistic scenario of Ref. [5], where possible. The values used were chosen to reflect hypothetical reference situations, thus ensuring the (reasonable) applicability of the calculated doses to worldwide situations.

The models described in Ref. [5] simulate the chemistry of a NORM residue deposit through a ‘lumped’ surrogate parameter (K_d). Therefore, the model used to estimate the doses is not capable of dealing specifically with detailed geochemical issues (for instance, acid generating heaps). However, as previously illustrated in Table 5, the estimated seepage concentrations based on the modelling in Ref. [5] compare well with the nominal concentrations inferred from measurements of various NORM residue deposits.

The doses calculated for each pathway included in this assessment are provided in Table 6. As can be seen, in a very sandy soil with low K_d values, it is possible that the dose to a child might marginally exceed 1 mSv/a. However, for more realistic, and still conservative, K_d values, the doses are well below 1 mSv/a.

The doses reported in Table 6 are based on the presence of both the ^{238}U and ^{232}Th decay series radionuclides in the residue deposit at an activity concentration of 1 Bq/g. The corresponding doses for the same pathways but with only the ^{238}U series radionuclides present in the residue deposit are provided in Table 7. The doses from only the ^{232}Th decay series radionuclides can be obtained by subtracting the doses in Table 7 from those in Table 6. As shown in Tables 6 and 7, the majority of the dose (>90%) is from the groundwater pathway.

TABLE 6. RESULTS OF PATHWAY CALCULATIONS FOR ^{238}U AND ^{232}Th DECAY SERIES RADIONUCLIDES, EACH AT 1 Bq/g

	Committed effective dose (mSv/a)			
	Realistic distribution coefficients		Sand distribution coefficients	
	Child	Adult	Child	Adult
Inhalation of dust	3.4×10^{-4}	6.5×10^{-4}	3.4×10^{-4}	6.5×10^{-4}
Ingestion of dust	0.011	0.0018	0.011	0.0018
External exposure	0.0056	0.0043	0.0050	0.0039
Ingestion of garden and agricultural products	5.2×10^{-5}	2.7×10^{-5}	2.8×10^{-4}	1.0×10^{-4}
Groundwater pathway	0.24	0.19	1.3	0.74
Surface water pathway (consumption of fish)	7.6×10^{-5}	3.9×10^{-5}	5.9×10^{-4}	2.5×10^{-4}
Total	0.26	0.20	1.3	0.75

TABLE 7. RESULTS OF PATHWAY CALCULATIONS FOR ONLY THE ^{238}U DECAY SERIES RADIONUCLIDES, EACH AT 1 Bq/g

	Committed effective dose (mSv/a)			
	Realistic distribution coefficients		Sand distribution coefficients	
	Child	Adult	Child	Adult
Inhalation of dust	1.3×10^{-4}	2.5×10^{-4}	1.3×10^{-4}	2.5×10^{-4}
Ingestion of dust	0.0070	0.0012	0.0070	0.0012
External exposure	0.0025	0.0019	0.0019	0.0015
Ingestion of garden and agricultural products	3.4×10^{-5}	2.1×10^{-5}	2.4×10^{-4}	9.4×10^{-5}
Groundwater pathway	0.16	0.15	1.1	0.67
Surface water pathway (consumption of fish)	4.7×10^{-5}	2.7×10^{-5}	5.5×10^{-4}	2.4×10^{-4}
Total	0.17	0.15	1.1	0.67

4. OVERALL CONCLUSIONS

From the doses reported in Tables 6 and 7, water related pathways are the largest contributor to dose with the majority of the dose (>90%) coming from the groundwater pathway. For the extremely conservative scenario with K_d values for sandy soils, there is some potential for the dose to a child to (slightly) exceed 1 mSv/a; however, for realistic K_d values, the doses to both the child and the adult receptors are well below 1 mSv/a.

In considering these doses, it is unreasonable, and extremely conservative, to consider direct consumption of undiluted leachate. In developing legislation for waste disposal, exposure scenarios have typically assumed that dilution by groundwater between the waste source and receptor will occur. Inert wastes or non-toxic wastes are defined in many jurisdictions (for example, the US Environmental Protection Agency (EPA) toxic leachate procedure) as wastes whose leachate has concentrations of elements <10 to 100 times higher than drinking water criteria [10]. As such, a reasonable exposure scenario would assume dilution and based upon precedent experience, a minimum dilution of 10–100 would be defensible.[3] In addition to dilution, natural attenuation and dispersion along with radioactive decay would reduce migration rates and concentrations; however, if the receptor water supply was close to the source, there would be no material radiological decay and the future concentrations would eventually attain the levels similar to the diluted groundwater. Since some of the NORM residue deposits will not be in equilibrium as a result of processing, it is useful to consider the radionuclides most likely to contribute to dose. This is illustrated in Table 8, which shows doses arising from the groundwater pathway by radionuclide. For a NORM residue deposit that only contains the radium (and associated decay products) from the uranium series (for example, a phosphogypsum residue deposit), the dose from the groundwater pathway would be less than 0.1 mSv/a using realistic distribution coefficients.

Moreover, in practice, it is unreasonable to assume that people would routinely drink an acidic water supply as it would not meet minimal drinking water quality standards for many conventional parameters, such as pH, TDS, iron and heavy metals, and aesthetics (taste and colour for example) and would be unpleasant to drink.

Although limited, available data summarized in Ref. [4] suggest that for actual situations, as opposed to hypothetical scenarios, the doses to people living near a NORM residue deposit containing uranium or thorium series radionuclide concentrations of 1 Bq/g are most likely to be below 1 mSv/a.

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[3] EPA originally proposed a dilution factor of 10 and later increased it to 100.

TABLE 8. RADIONUCLIDE-SPECIFIC DOSES FROM THE GROUNDWATER PATHWAY

	Dose, mSv/a			
	Realistic distribution coefficients		Sand distribution coefficients	
	Child	Adult	Child	Adult
U	0.071	0.093	0.10	0.13
^{230}Th	0.0057	0.010	0.0018	0.0032
^{226}Ra	0.011	0.011	0.027	0.027
^{210}Pb	0.0099	0.0067	0.18[1]	0.12
^{210}Po	0.061	0.029	0.081	0.039
^{228}Th	0.014	0.0066	0.0044	0.0021
^{232}Th	0.0062	0.011	0.0019	0.0035
^{228}Ra	0.063	0.027	0.16	0.067
Total	0.24	0.19	1.3	0.74
Total (only ^{238}U series)	0.16	0.15	1.1	0.67
Total (only ^{226}Ra, ^{210}Pb, ^{210}Po)	0.081	0.046	1.0	0.54

In view of the doses calculated for the representative NORM residue deposit and other available information as summarized in this paper, although there is a possibility that doses slightly in excess of 1 mSv/a might occur in unusual circumstances, it is considered more likely than not that the dose to an adult or child who lives in close proximity to a NORM residue deposit would be well below 1 mSv/a for the vast majority of ‘real’ situations.

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REGULATORY GUIDELINES FOR USE OF PHOSPHOGYPSUM IN BUILDING AND CONSTRUCTION MATERIALS IN INDIA

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Abstract

Rock phosphates used for the production of phosphoric acid in India contain moderate concentrations of radionuclides of natural origin, such as ^{238}U and ^{226}Ra. Processing of rock phosphates for the production of phosphoric acid by the wet process (sulphuric acid attack) results in most of the uranium going to the phosphoric acid and most of the radium going to the by-product phosphogypsum. The phosphogypsum so produced can be used in various commercial applications such as in building and construction materials. A radiological impact assessment for use of phosphogypsum in building and construction materials was carried out by the Atomic Energy Regulatory Board (AERB) of India, which is the national nuclear regulatory body. The study comprised the collection of nationwide data on the production of phosphogypsum, an analysis of the ^{226}Ra content of the phosphogypsum samples, field visits to industries manufacturing various construction materials such as plaster of Paris, cement, plaster boards and panels. Based on the dose estimation for various postulated scenarios and international practices, and taking socio-economic factors into account, regulatory guidelines have been formulated by the AERB for the use of phosphogypsum in building and construction materials in India. The paper discusses the basis for and details of the development of these regulatory guidelines.

1. INTRODUCTION

Fertilizer plants in India are presently processing rock phosphates imported from various countries, including Jordan, Morocco, Egypt, Senegal, Togo and China, for the production of phosphoric acid and fertilizer. With 12 operating plants presently in the country, India is one of the top four countries in the world in the production of phosphatic fertilizers. Rock phosphates are known to contain enhanced concentrations of radionuclides of natural origin, especially uranium and its daughter products such as ^{226}Ra. In the chemical processing of rock phosphates, about 0.3 t of phosphoric acid and 1.5 t of the major by-product (or solid waste) phosphogypsum is produced per tonne of rock phosphate and the radionuclides become selectively separated into the acid and solid waste [1]. The radionuclides of significance in the phosphogypsum are ^{226}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po. Besides these radionuclides, phosphogypsum also contains residual acidity, fluoride compounds and toxic trace metals such as Cd, Pb, Cr and As. The phosphogypsum so produced is used in various commercial applications, one among them being building and construction materials. A radiological impact assessment for the use of phosphogypsum in building and construction materials was carried out by the Atomic Energy Regulatory Board (AERB) of India, which is the national regulatory body to ensure radiological safety in the country.

2. REVIEW OF INTERNATIONAL REGULATIONS AND PRACTICES ON THE USE OF PHOSPHOGYPSUM

In order to carry out a dose assessment for the use of phosphogypsum in building and construction materials, a review of international regulations and practices in various countries was carried out. It was observed that in the United States of America, current Environmental Protection Agency regulations do not permit the use of phosphogypsum above 0.37 Bq/g for any agricultural or industrial applications. Some countries like Germany have stopped manufacturing phosphoric acid and depend on imports to meet their needs. Some industries in the Netherlands obtained permission for the use of phosphogypsum for building purposes following a risk assessment route. Australia and South Africa have started working to evolve guidelines to address immediate problems of the industry. In the final framework of the Australian National Directory of Radiation Protection, there are proposals to include items such as exclusion from regulation of NORM at less than 1 Bq/g (for the head of chain radionuclide), conditional exemption from regulation in circumstances where protection is optimized and where individual doses are less than about 1 mSv/a and the application of a graded approach (commensurate with the extent of the hazards involved) should regulatory control need to be applied. Various international guidance documents relevant to the regulation of phosphogypsum are listed below.

(a) IAEA Safety Series No. 115 (1996) specifies an exemption level of 10 Bq/g for ^{226}Ra, based on individual dose of 10 μSv/a, but this exemption level is not applicable to bulk quantities of material [2].

(b) The European Commission report RP112 on Radiological Protection Principles concerning the Natural Radioactivity of Building Materials (1999) recommends an incremental effective dose of 0.3 mSv/a as the criterion for exemption and clearance [3]. This criterion has been justified with the following arguments:

(i) It is comparable with regional variations in dose from natural background radiation.

(ii) It is coherent with dose constraints recommended by the International Commission on Radiological Protection (ICRP) for effluents (0.3 mSv/a for the nuclear industry).

(iii) It is below the lower marker point for worker exposure in 'work activities' (a European Commission term for non-nuclear industries).

(iv) RP 112 recommends, for building materials, that doses above 1 mSv/a should be accepted only in some very exceptional cases and that controls can be based on a lower dose criterion if it is judged that it is desirable and will not lead to impractical controls. It is therefore recommended that controls should be based on a dose in the range of 0.3 to 1 mSv/a.

(c) The European Commission report RP122 on Practical Use of the Concepts of Clearance and Exemption – Part II (2001) also indicates a dose increment of 0.3 mSv/a for exemption and clearance in connection with 'work activities' [4].

(d) IAEA Safety Standards Series No. RS-G-1.7, Application of the Concepts of Exclusion, Exemption and Clearance (2004) recommends an activity concentration of 1 Bq/g for individual uranium and thorium series radionuclides as an entry point for the regulation of bulk quantities of material (based on a consideration of the worldwide distribution of activity concentrations for these radionuclides) [5]. This guidance is elaborated on in IAEA Safety Reports Series No.49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials (2006) [6]. Below 1 Bq/g for individual uranium and thorium series radionuclides it is usually unnecessary to regulate irrespective of the quantity of material or whether it is in its natural state or has been subjected to some form of processing. However, the use of building materials with activity concentrations below these values may need some regulatory consideration.

3. INFORMATION COLLECTED THROUGH QUESTIONNAIRE

A questionnaire was prepared and sent to all the operating phosphate fertilizer plants, seeking information such as the location of the phosphogypsum disposal sites, the amount of phosphogypsum stored and sold, the various uses of phosphogypsum including construction materials and the radiometric analysis of phosphogypsum. In reviewing the responses to this questionnaire, it was noted that:

(a) Phosphogypsum is presently disposed of at designated disposal sites near the plants.
(b) Residential areas extend to within 1–2 km of the phosphogypsum disposal sites. The phosphogypsum disposal sites are 0.5–6 km from the nearest surface water source. Runoff from the phosphogypsum disposal sites is mostly collected and recycled.
(c) About 60% of the phosphogypsum produced is sold for use in various commercial applications. It is estimated that about 70% of the material sold is used for cement manufacture where it is blended with clinker as an additive to retard the setting time in accordance with the designated grade (resulting in a phosphogypsum content of 2–5%). Presently, only a small portion of the phosphogypsum is used in the manufacture of other building materials. In the manufacture of plaster of Paris, a mix of about 80% phosphogypsum to 20% chalk is used. In the manufacture of plasterboard, a mix of about 30% phosphogypsum and about 70% natural gypsum, together with small quantities of additives, is used. Phosphogypsum is also being considered by many fertilizer plants for the manufacture of glass fibre reinforced gypsum panels.
(d) Phosphogypsum produced in the fertilizer plants in India using imported rock phosphate contains ^{238}U and ^{226}Ra at typical activity concentrations of 0.1–0.2 and 0.5–1.3 Bq/g, respectively.

4. FIELD VISITS AND RADIOLOGICAL MONITORING

Visits were made to a phosphate fertilizer plant where the phosphogypsum is produced and stored and also to one of the facilities where phosphogypsum is used for the manufacture of plaster of Paris and plasterboard. The exposure rates measured at these locations are summarized in Table 1. The natural background radiation level in the surrounding areas was 0.03–0.06 μGy/h.

Samples of stucco, plaster of Paris and plasterboards were collected and analysed for radioactivity. The results of the analyses are given in Table 2.

5. DOSE ASSESSMENT

Various scenarios were postulated and a dose assessment for each scenario was carried out using methodologies reported for the assessment of radiation

TABLE 1. EXPOSURE RATES IN PHOSPHATE INDUSTRY FACILITIES

	Absorbed dose rate (μGy/h)
Phosphate fertilizer plant:	
Grinding section	0.05–0.07
Filtration section	0.22–0.24
Phosphogypsum disposal area, at 1 m	0.22–0.24
Phosphogypsum disposal area, on contact	0.27–0.35
Plaster of Paris plant:	
Feed material on conveyor belt	0.08–0.09
Dried feed (after rotary drier)	0.08
Calciner area	0.04
Product storage area, at 1 m	0.08–0.1
Product storage area, on contact	0.17
Plasterboard plant:	
Silo bottom (mixer of natural gypsum and phosphogypsum)	0.04
Hammer mill	0.04
Tensioner	0.03
Aligner	0.03
Mixer	0.03
Product stacking area	0.09–0.1

TABLE 2. ACTIVITY CONCENTRATIONS IN PHOSPHOGYPSUM PRODUCTS [7]

	Activity concentration (Bq/g)			
	^{238}U	^{226}Ra	^{232}Th	^{40}K
1.25 cm plasterboard	0.009 ± 0.0011	0.0924 ± 0.0027	0.0048 ± 0.0004	0.0153 ± 0.001
Stucco	0.0874 ± 0.0073	0.338 ± 0.006	0.0106 ± 0.001	0.0072 ± 0.001
Plaster of Paris	0.109 ± 0.0122	0.383 ± 0.0115	0.0135 ± 0.0021	0.0488 ± 0.001

exposures from building materials containing radionuclides. These methods make use of validated theoretical models. The internal exposure component was not considered, since the release of radon and consequent exposure will not be significant in Indian conditions, mainly because of once-through ventilation conditions [8].

Scenario 1: Walls made of phosphogypsum bricks and cement containing phosphogypsum

The exposure inside a 5 × 4 × 2.8 m room with walls 30 cm thick has been estimated [9]. The use of phosphogypsum based materials (0.629–1.08 Bq/g ^{226}Ra) for construction of the walls resulted in an increase in external dose rate of 0.164–0.211 μSv/h over that for walls constructed of normal clay bricks and natural gypsum based cement (0.021–0.048 Bq/g ^{226}Ra). In the highly unlikely situation of an individual living in the room for 7000 h/a, the incremental dose would be 1.4 mSv.

Scenario 2: Normal cement walls lined with phosphogypsum plasterboard

In this scenario, the walls and ceiling of a 5 × 5 × 3 m room are lined with 1 cm thick phosphogypsum plaster boards containing ^{226}Ra at an activity concentration of 0.4 Bq/g. It has been estimated that the dose from external exposure received by a person continually occupying the room would not exceed about 0.13 mSv/a [10]. When considering 1.25 cm plasterboard (as maufacured in India) containing 30% phosphogypsum with a ^{226}Ra activity concentration of 1.3 Bq/g (the maximum concentration noted in phosphogypsum), the dose from external exposure would be 0.15mSv/a.

Scenario 3: Floor, ceiling and walls made of phosphogypsum panels

In this scenario, a 5 × 4 × 2.8 m room is constructed from compact phosphogypsum panels with a thickness of 20 cm, a density of 2.35 g/cm^3 (such that 1 m^2 of wall weighs 470 kg) and a ^{226}Ra activity concentration of 1 Bq/g [4]. It has been estimated that a person living in the room for 7000 h/a would receive an annual dose from external exposure of 4.5 mSv. However, when considering hollow phosphogypsum panels (such that 1 m^2 of wall of the same thickness weighs only 38 kg) with a ^{226}Ra concentration of 1.3 Bq/g (the maximum noted in phosphogypsum), the corresponding annual dose for 7000 h occupancy would be about 0.46 mSv.

Since cement manufacturers use only 2–5% of phosphogypsum, the contribution to exposure from this source will be minimal. For uses such as plaster of Paris and road base material, the exposures are likely to be much lower than in the above scenarios.

6. FORMULATION OF REGULATORY GUIDELINES

There are proposals from fertilizer plants for constructing standard glass fibre reinforced phosphogypsum panels typically of the size 12 × 3 m and 12.5 cm thick, with 48 hollow cavities. These panels will be used for constructing buildings under the poverty alleviation scheme. In future, there may be proposals for other dimensions as well. It was estimated that for compact panels, doses can be as high as 4.5 mSv/a. Hence, considering the radiological impact as well as other socio-economic factors, it was decided to have a regulatory limit on surface activity and to restrict the concentration of ^{226}Ra in the phopshogypsum to 1 Bq/g as given in IAEA Safety Guide-RS-G-1.7 [5] such that the radiological dose to the inhabitant of a building constructed from phosphogypsum panels is limited to 0.3 mSv/a.

A draft safety directive specifying various guidelines was prepared, circulated to stakeholders for comment, and then finalized as follows:

> “AERB approval is not required for selling phosphogypsum for its use in building and construction materials provided the activity concentration of Ra-226 in it is less than or equal to 1 Bq/g. [If Ra-226 concentration in phosphogypsum is more than 1Bq/g, it is to be mixed with other ingredients such that the Ra-226 activity concentration in bulk material is less than or equal to 1.0 Bq/g.]
>
> “AERB approval is not required for manufacturing and use of phosphogypsum panels or blocks provided they have Ra-226 activity less than 40 kBq/square metre area of any surface of the panels/blocks.”

7. CONCLUSIONS

The surface density of the panel is regularly measured by the panel manufacturers as part of their quality control. The surface activity of ^{226}Ra per square metre of the panel surface can be estimated from the product of the activity concentration of ^{226}Ra in phosphogypsum and the surface density. To assess the activity concentration of ^{226}Ra in phosphogypsum, all phosphate fertilizer plants have been directed by AERB to carry out analyses to determine the ^{238}U and ^{226}Ra content in the phosphogypsum produced from the processing of each imported consignment of phosphate rock and to report the results to AERB on a quarterly basis. These data are being periodically reviewed in order to decide on the frequency of such analysis in future.

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IDENTIFICATION OF NORM FACILITIES IN BULGARIA — METHODICAL APPROACH AND RESULTS

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Abstract

Bulgaria has been a member of the European Union since 2007. Consequently, the European standards and regulatory framework have to be implemented in the Bulgarian legislative system. As part of this process, the basic safety standards in the field of radiation protection (EU BSS), as laid down in the Council Directive 96/29/Euratom, have to be implemented. Title VII of the EU BSS concerns natural radiation sources and related work activities. Its implementation requires that each Member State ensure the identification of work activities that may be of concern regarding naturally occurring radioactivity. This identification has to be done by surveys or other appropriate methods. Guidance concerning the methods of such investigations is given in the recommendations contained in the European Commission report RP88, as well as in the pertinent literature describing the occurrence of radioactivity in industrial facilities. But practical experience has shown that a comprehensive overview of the radioactivity of NORM or TENORM in an individual facility is difficult to obtain in a single survey. Furthermore, even if an industry is not radiologically relevant today, it may become so after changes of feed materials, as well as changed or new technologies (for example, installation of dust filters). This necessitates an approach that takes into account the processes that may lead to the formation of NORM and thus allows the anticipation of the occurrence of radiologically relevant materials in the future. In order to comply with the requirements of Title VII of the EU BSS, desktop research and field investigations were carried out and the evaluated, the results of which were used to enable the Bulgarian Nuclear Regulatory Authority to complete the surveys and obtain a comprehensive picture of the situation concerning NORM industries in Bulgaria. Based on the findings in the field, a regulatory framework to address work activities and materials involving natural radioactivity was developed also. In the paper, the methodical approach for the survey and the results obtained are described. The results of this investigation have allowed the competent authority to decide on the level of the necessary regulatory regime of NORM related activities in Bulgaria. The method has turned out to be practicable and can be applied to regions or countries in which a systematic investigation of naturally occurring radioactivity in industrial sectors is necessary.

1. INTRODUCTION

Bulgaria, in the Balkans in south-eastern Europe, covers an area of 111 910 km^2 with a population of 7.6 million. In 2007 Bulgaria became a Member State of the European Union. Currently, there are no specific regulations to detail the general requirements related to radiological safety of NORM activities in Bulgaria. In order to assist with the implementation of Title VII (“Significant increase in exposure due to natural radiation sources”) of the basic safety standards in the field of radiation protection (EU BSS), as laid down in the Council Directive 96/29/Euratom [1], into Bulgarian legislation, a project funded from the PHARE programme of the European Commission (EC) was launched. This project, the beneficiary of which was the Bulgarian Nuclear Regulatory Agency (BNRA), was aimed at identifying work activities involving natural radiation sources and NORM materials on a national level, which require regulatory attention, and to develop a regulatory framework for such work activities and materials. According to Article 40 of the EU BSS, the identification of work activities that may be of concern regarding naturally occurring radioactivity has to be done by surveys or other appropriate methods.

The methodical approach applied for this purpose included the following steps:

(1) Pre-check of industry sectors based on the list of industry sectors from the IAEA [3] and the ‘positive list’ of the EC [4];
(2) Identification of waste types of potential concern from a process analysis;
(3) Selection of available and accessible facilities or sites and preparations for a fact finding mission;
(4) Fact finding mission with in-situ measurements and sampling;
(5) Evaluation of results with exposure estimations and conclusions regarding a national positive list.

Based on the results of the surveys and an analysis of the existing Bulgarian legislation, a new regulatory document for work activities and residues was drafted. The project was successfully completed in December 2009 [2].

2. SURVEYS

Surveys should enable a screening of the sites and industrial activities that may require further and detailed investigations. The methodology of the site investigations and the assessments of the results should be in line with the concepts of exemption and clearance.

As a first step, a preliminary list of industries was developed, which may be relevant in the context of NORM or TENORM. This list was based on a ‘macro-economic approach’, that is, using national industry statistics and other publicly available information on the Bulgarian economy. This information was checked against the list published by the IAEA and the ‘positive list’ proposed by the EC and served as a first selection criterion for the size and relevance of the industries that may be investigated. Soon, however, it became clear that due to the structural changes of the Bulgarian economy over the last two decades a significant part of the former industries in Bulgaria had gone out of operation. These changes affected in particular the resource and basic industries, namely those that are most likely to need regulation. For instance, the only Bulgarian pig iron producer near Sofia was shut down a few years ago.

As the project duration was restricted to one year, the goal of a comprehensive survey of the national situation concerning NORM related work activities and residues would be illusionary. Rather, the survey had to be restricted to exemplary investigations, which provide the methodology for further investigations and demonstrate the general relevance of a given industrial sector for radiation protection in the country.

In preparation for the site visits, practical issues such as the availability of management representatives to grant access and logistical accessibility played a decisive role. The selection of the sites of intended visits was therefore an iterative process involving the competent authority (BNRA), the consultants and the respective plant operators. Table 1 lists the sites which were selected for exemplary investigations.

Even though uranium mining and milling is not commonly regarded as a NORM industry but is typically seen as a licensed practice as part of the nuclear fuel cycle, some uranium mining and milling sites such as Eleshnitsa were also included in the survey, due to the radiological and technical similarities with more conventional NORM sites.

TABLE 1. SITES SELECTED FOR EXEMPLARY INVESTIGATIONS

Site	Industry
Pernik/Blagoevgrad	Drinking water utility
Eleshnitsa	Former uranium mine, treatment of mine effluent
Mezdra	Ceramics production using zircon and zirconia
Varna (Agropolihim)	Phosphate fertilizer production
Maritsa-Radnevo	Coal fired power plant
Asarel-Medet, Panagyurishte	Copper ore mining and smelting
Ihtiman	Production of thoriated welding rods
Momin Prohod Hisarya, Narechenski Bani	Mineral water springs and spas
Dolni Dabnik, Pleven	Oil and gas production

3. FACT FINDING MISSION

Following the selection of exemplary sites to be investigated, the site visits were planned in the following way:

(a) Detailed ‘industry fact sheets’ were prepared and sent in advance to the site operators. They contained information about the technological processes that typically lead to the formation of NORM, the types of radiation (γ, β) encountered and a motivation letter describing why radiation protection may be an issue in the industry.

(b) As a methodical tool for the inspection teams, checklists and instructions regarding on-site sampling equipment, sample storage and transportation (solid, liquid), use and calibration of radiation measuring devices, as well as health and safety instructions were prepared.

Further hints concerning the methods of such investigations are given in the European Commission report RP88 [5], as well as numerous papers and guidelines [6] describing the occurrence of radioactivity in industrial facilities. The fact finding mission was carried out in March and April 2009. Site inspection teams consisted of the consultant and representatives of the competent authority, as well as the site operator, typically the health and safety officer. It was notable that the awareness of NORM issues among the industry representatives was very low, even in industries which are widely known for the occurrence of NORM. At the sites, measurements were made at the relevant points of the technological processes, and solid and liquid samples were taken where material was available.

All samples were radiochemically analysed by a laboratory certified according to ISO 17025.

The experience gained in several years of NORM surveillance, including a predecessor project with similar objectives in Romania in 2008, had shown that the identification of sites and facilities which are typically contained in the positive lists is not sufficient to draw conclusions on the need for regulatory control of that industry as a whole, and may even be misleading. Therefore, the following supplementary aspects must be taken into consideration:

(1) A plant may not be operational at the time of the investigation, but temporarily shut down for maintenance. Raw materials and residues may therefore not be available for sampling.
(2) Raw materials used at the time of the site visit may coincidentally not be representative. In particular, their radiological properties may be untypical.
(3) Raw materials and processes may change. For example, a phosphate fertilizer plant may import raw phosphate from different sources with varying radionuclide concentrations. Dust filters may be installed in the future, which minimize airborne discharge of dust but necessitate the management of filter dust rich in ^{210}Pb and/or ^{210}Po.
(4) The level of radioactivity in the raw materials may vary significantly between individual sites. This is particularly true for mineral water springs and spas, and natural oil or gas production sites. Therefore, even if a particular site shows no elevated level of radioactivity, this is not sufficient to draw a firm conclusion for the entire industrial sector in the country.

Moreover, the usual positive lists of NORM-industries are very general and do not give sufficient advice regarding the degree of radionuclide enrichment in an individual process stream of a certain facility. These problems may at least be partially overcome using a process-based approach as described in Ref. [7], for example. It gives the opportunity to:

(i) Identify the relevant parts of an operation based on general physical or chemical considerations;

(ii) Select the appropriate radiation detectors (for example, beta sensitive devices if furnace dust dominated by ^{210}Pb is expected);
(iii) Give detailed instructions to the laboratory to which samples are sent, to use appropriate spectrometry equipment (for example, to take account of the 46 keV energy peak for ^{210}Pb);
(iv) Draw conclusions on the age of residues (for example, from the activity ratio of ^{228}Ra and ^{228}Th).

4. RESULTS OF THE FACT FINDING MISSION

The data collected during the site investigations, partly complemented by information from the available literature, were used to carry out dose assessments for workers and members of the public. As no binding dose assessment guidelines are presently available in Bulgaria, the German guidelines [8] were used, having proven to be rather practicable. However, it must be noted that the dose estimates are sometimes beset with great uncertainties. Therefore, assumptions have been made and model parameters have been used which reflect a likely exposure scenario or, if no information was available, describe a conservative approach. As a general rule, the following approach was applied to the dose calculations:

(a) If the site visits have revealed activity concentrations and other radiologically relevant material properties which are comparable with international literature data, and if consequently they lead to exposure of members of the public or workers which cannot be neglected from the radiation protection point of view, the results of the dose calculations are taken as the basis for the assessment.

(b) If, for any reason (such as those mentioned above) the dose estimates do not lead to any elevated exposures at the existinglocation, values of the radiological properties from the literature have been used for hypothetical scenarios which may happen in the future. In this case, the scenario for the existing situation and the hypothetical scenario for future activities are both taken into account for deriving the conclusions on the radiological relevance of a particular work activity or material.

While raw data from individual sites are usually treated with confidentiality, the aggregated results are summarized in Table 2. The dose estimates have revealed the following facts:

(1) The overall level of natural radioactivity, which is not particularly high compared with other countries in Europe [9], is reflected in a small number of sites and industries that deserve immediate attention.

(2) Industries processing raw materials that originate in Bulgaria are unlikely to lead to significantly elevated levels of radioactivity in their residues or other process streams. However, if raw materials are imported, such as phosphate rock from deposits known for their radiological relevance, regulatory oversight may be warranted.

(3) Some industries that were not operational during the project implementation may be revived when economic conditions improve. Repeat measurements should then be carried out to confirm or dispel radiological concerns.

Of the industries listed in Table 1, only the following require immediate regulatory attention:

(i) Phosphate fertilizer production;
(ii) Use of thoriated products such as welding rods;
(iii) Geothermal and mineral water, following a more detailed country-wide survey;
(iv) Oil and gas production.

In the preparation of a regulatory framework for NORM related work activities and residues, however, it was decided to use a rather broad positive list containing most of the elements of the draft revision of the EU BSS [4], in order to also cover industries which may become radiologically relevant in the future even if they are presently not operating or do not lead to elevated effective doses today but may do so with changed technologies and/or raw materials. This broad approach also ensures compatibility with the anticipated changes that are being made in the process of revising the EU BSS.

5. CONCLUSIONS

The survey has shown that Bulgaria is a region of low background radioactivity, owing to its geology in large parts of the country. The limited number of sites available for the surveys makes it difficult to identify those work activities that are really of concern regarding naturally occurring radioactivity in Bulgaria in a comprehensive fashion, but allows one to conclude that the number of industries and sites requiring regulatory attention is limited to a small number. With expected economic growth and possible changes in feedstocks and/or technologies of NORM industries in Bulgaria, a positive list restricted to the current situation seems inadequate. Therefore, a positive list has been developed that includes a broad summary of potential NORM industries of potential concern in Bulgaria. This ensures compatibility with the positive list of the draft revision of the EU BSS [4]. Furthermore, the proposed positive list supports the administrative process by specifying the industrial processes and materials that need regulatory attention.

In summary, the investigation has provided an overall picture of the Bulgarian situation with respect to NORM and has formed the basis for deriving the level of the necessary regulatory regime for work activities and materials involving NORM in Bulgaria. The methodology chosen has turned out to be practicable and adequate. It can be applied to other regions or countries that require a systematic investigation of naturally occurring radioactivity in their industries.

TABLE 2. RESULTS OF DOSE ESTIMATES

Industry, process	Radiological relevance		Remarks
	Workers	Members of the public	
Water treatment	Currently no problems known, but doses may come close to 1 mSv/a	No significant doses except if wastes are used in construction materials	If residues such as filter sands occur, their use must be restricted
Water treatment at former uranium mining and processing site	Dose <1 mSv/a, except for ^{222}Rn in treatment facilities which may lead to doses exceeding 1 mSv/a	Doses <1 mSv/a	Uranium mining is not commonly regarded as a NORM industry, but part of the nuclear fuel cycle
Ceramics industry	Negligible	No realistic exposure scenarios	
Phosphate fertilizer industry	Doses may significantly exceed 1 mSv/a	Groundwater pathway may lead to significant doses and must be investigated in detail	Only one company active but currently shut down. Use of residues for construction materials must be restricted
Coal-fired power plants	Dust can lead to exposure >1 mSv/a, but in general doses <1 mSv/a	No significant doses except if wastes are used in construction materials	Use of residues for construction materials must be restricted
Ore mining	No site known at the moment where dose may exceed 1 mSv/a, but may become an issue in the future. Radon-222 may become a source of exposure	Doses typically <1 mSv/a	
Thoriated welding rods and other products	Doses may significantly exceed 1 mSv/a	Not relevant	
Geothermal water	Doses <1 mSv/a for conservative assumptions, but depend on work conditions	Doses may exceed 1 mSv/a	

TABLE 2. RESULTS OF DOSE ESTIMATES (cont.)

Industry, process	Radiological relevance		Remarks
	Workers	Members of the public	
Metal smelters	Doses <1 mSv/a	Uncontrolled spread of filter dust may lead to doses >1 mSv/a	Uncontrolled spread of filter dust must be precluded (car and tyre wash plants!)
Oil and gas production	Doses may significantly exceed 1 mSv/a	No exposure scenarios leading to doses >1 mSv/a	

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EUROPEAN WASTE CATALOGUE — A PLATFORM FOR A COMMON APPROACH TO NORM AND OTHER INDUSTRIAL WASTE

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Abstract

The European Council Directive 96/29/Euratom requires European Union (EU) Member States to determine which NORM 'work activities' are of concern with respect to radiation exposure. Existing data have highlighted a strong need to include, within the regulatory scope of radiation protection, non-nuclear industries in which materials containing enhanced natural radioactivity occur. In reality, however, these radiation risks are often still not subject to control and are not even subject to simple monitoring. This is brought about by a lack of information about radiation risks outside those industrial sectors in which sources of radiation are purposely used (practices). As a result, the potential end users of NORM type residues are seldom aware of possible problems due to natural radioactivity, or are afraid of the negative consequences (such as costs, and fears among workers and the public) should radiation protection measures be implemented. Finally, the radiation risk caused by the use or disposal of NORM is in many cases neglected. EU Member States tend to deal with this problem in a case by case manner, but it is still rare for the risk caused by NORM to be fully taken into account when the destination of such a residue is planned. Often, the treatment of NORM residues is regulated only by 'classical' environmental regulations, which take into account only the non-radiological risks. This may engender serious problems from a radiation protection point of view. An approach to the solution of the problem of NORM, based on the application of universal so-called 'clearance levels' is not always appropriate and, often, case specific risk assessment scenarios must rather be developed. The paper promotes the idea of using the European Waste Catalogue (EWC) to help develop an awareness among all stakeholders of concern. A list of waste types already classified in the EWC has been completed with basic information about the possible content of natural radioactivity. A form for additional information to support a derived radiation risk assessment is proposed. As an additional advantage, attention is drawn to the role of the EWC as a tool for harmonization and consistency in the approach to dealing with the radiological and chemical risks associated with NORM residues.

1. INTRODUCTION

Since radiation risk is usually considered to be related to nuclear energy, most research on radiation protection has focused on radionuclides of artificial origin in radioactive wastes, spent nuclear fuel or global fallout caused by atomic bomb tests and nuclear power plant failures. Far less attention has been paid to the radiation risk caused by exposure to ionizing radiation originating from natural sources. Apart from radon, radiation emitted by primordial radionuclides in their natural state (unaltered by human activity) is not considered to be a source of risk, whether to human beings or to the environment. There are many areas in the world having an elevated content of naturally radioactive elements caused either by the geological and geochemical structure of the rocks, or by the radioactive content of water flowing from underground springs [1]. However, if concentrations of radionuclides of natural origin have been changed, deliberately or accidentally, as a consequence of industrial activity, it is quite another matter. According to current radiation protection principles, the related risk, excluding the influence of the so-called natural background, must be assessed in a similar manner as the risk caused by artificial sources.

Technologically enhanced naturally occurring radioactive materials (TENORM) are common in many types of non-nuclear industry. Many processes within such types of industry lead to a situation where the activity concentrations of radionuclides of natural origin are enhanced. Such situations may take place, for example, in industrial processes where significant mass reductions of raw materials occur. Usually, the radionuclides accumulate in waste. Such alterations to the natural state can result in an incremental radiation risk to people as well as to the environment. Each particular occurrence of natural radioactivity presents a unique scenario of exposure — usually different from those caused by artificial radionuclides present in radioactive waste or spent nuclear fuel. Frequently, the amounts of such types of waste can be up to hundreds of thousands of cubic meters or tonnes and they are often placed directly into the environment. In the coal mining industry, the radium activity deposited in single tailing ponds may reach 300 GBq [2]. Probably, the biggest ‘producers’ of waste with enhanced concentrations of radionuclides of natural origin are phosphate processing plants, where radionuclides remain within the phosphogypsum that is subsequently stored in disposal sites near the plants at a rate reaching 350 MBq/h [3]. In spite of this, the risks associated with natural radioactivity have been discussed mainly for areas with elevated natural radiation, as instances where the effects of low doses can be studied (see, for instance, Refs [4, 5]. Until relatively recently, the enhanced natural radiation resulting from the non-nuclear industry has not been the focus of radiation protection interest.

2. LEGISLATIVE ASPECTS OF NORM

At the European Community level, the importance of the radiation risk caused by natural radioactivity was first reflected in legislation in the Council Directive 96/29/Euratom [6]. In paragraph 40 of this directive, it is stated that:

“1. This Title shall apply to work activities not covered by Article 2 (1) within which the presence of natural radiation sources leads to a significant increase in the exposure of workers or of members of the public which cannot be disregarded from the radiation protection point of view.

2. Each Member State shall ensure the identification, by means of surveys or by any other appropriate means, of work activities which may be of concern. These include, in particular:
   - (a) work activities where workers and, where appropriate, members of the public and exposed to thoron or radon daughters or gamma radiation or any other exposure in workplaces such as spas, caves, mines, underground workplaces and aboveground workplaces in identified areas
   - (b) work activities involving operations with, and storage of, materials, not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of workers, and, where appropriate, members of the public
   - (c) work activities which lead to the production of residues not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of members of the public, and, where appropriate workers
   - (d) aircraft operation.”

The Directive left the crucial decision about the significance of risk caused by natural radioactivity to the competency of each Member State. Faced with the lack of well founded information about NORM and TENORM, many Member States introduced into their own regulations only those cases mentioned explicitly in the Directive, namely underground mining, caves and spas. For these countries, the problems of waste containing enhanced concentrations of natural radioactivity are mostly outside the scope of any regulation.

In this light, the most generic problem is identification of circumstances where enhanced natural radioactivity can cause significant risk. There are different national approaches to what constitutes NORM and TENORM and, consequently, to which industries wastes and products are covered by relevant guides and regulation. In each EU Member State, the national radiation safety legislation establishes a uniform legal framework for regulatory control of practices. Practices are related to the deliberate use of ionizing radiation, radioactive sources or fissile materials. These practices are carried out under predetermined circumstances and the associated parameters such as source activity, radiation type and dose rate are well defined. In such cases, the monitoring of parameters determining radiation risk is obvious. Regulatory control is applied when the radiation risk exceeds specific criteria such as exemption levels.

Such an approach is well founded and effective only in cases where specific control of the radiation risk is assured or at least can be assumed. Otherwise a vicious circle can appear: there is no information about the radiation risk in the absence of controls, so there is no reason to implement regulatory control. Thus there is no regulatory control and no driving force to embark on any activity focused on evaluation of the radiation risk. Ultimately, even a severe radiation risk can be overlooked.

In the case of radiation risk caused by enhanced natural radioactivity, the necessity for regulatory control is not obvious. Natural radioactivity is a primordial property of matter surrounding us. Radionuclides of natural origin are present in almost all substances that we deal with. Therefore, the generic question is: When is their presence significant from a radiation protection point of view? The answer seems to be very simple: When the derived radiation risk exceeds an acceptable level, that is, when the necessity for regulatory control is apparent. But to obtain such an answer is not so simple. In the case of exposure to the risk caused by natural radioactivity, there are usually no specific controls in place in the absence of precise regulation. This results in a knowledge gap about the real radiation risk. There is no knowledge as to whether allowable limits of risk have been exceeded and therefore there is no consideration of the need for regulatory control. This implies that there is no risk evaluation and no need for control. In this way, even serious levels of radiation risk may remain outside the scope of application of radiation protection control measures.

Additional complexity is introduced into the problem by a different and sometimes conflicting regulatory framework for radiological and non-radiological aspects, including the prescribed waste management routes. Ionizing radiation, because of its carcinogenic and mutagenic properties, could be considered as implicitly included in environmental regulations (regulations other than those dedicated specifically to radiation protection) but the environmental authorities generally defer to radiation protection authorities for the specific aspects and the vicious circle is closing again. Finally, the hazard caused by radiation originating from naturally radioactive materials is rarely taken into consideration when the treatment of industrial waste is planned and an environmental risk assessment is carried out. There is a great need to provide the non-nuclear industry operators with well founded information about the real risk caused by enhanced natural radioactivity.

3. CURRENT SOLUTION

The solution applied in many countries consists of identifying the industrial activities for which the presence of natural radioactivity could cause a significant radiation risk based on available data. An IAEA Safety Report lists, in addition to uranium mining and processing, 11 types of industrial activity [7]. The so-called ‘positive list’ now being proposed by the European Commission for incorporation into a revised European Council Directive [8] identifies 14 types of work activity, whereas in the current European Directive [6] only a limited number of examples are listed, such as spas, caves and underground mining. The IAEA list and the proposed European ‘positive list’ are slightly different, mainly because of differences in the way in which specific work activities are demarcated. Details are given in Tables 1 and 2, along with identification codes that have been assigned for the purposes of this paper.

TABLE 1. INDUSTRY SECTORS IDENTIFIED IN REF. [7] AS BEING THE MOST LIKELY TO REQUIRE SOME FORM OF REGULATORY CONTROL

	Code assigned for purposes of this paper
Extraction of rare earth elements	IAEA 1
Production and use of thorium and its compounds	IAEA 2
Production of niobium and ferro-niobium	IAEA 3
Mining of ores other than uranium ore	IAEA 4
Production of oil and gas	IAEA 5
Manufacture of titanium dioxide pigments	IAEA 6
The phosphate industry	IAEA 7
The zircon and zirconia industries	IAEA 8
Production of tin, copper, aluminium, zinc, lead, and iron and steel	IAEA 9
Combustion of coal	IAEA 10
Water treatment	IAEA 11

Note: The industry sectors listed are those identified in addition to the mining and processing of uranium ore.

TABLE 2. ‘POSITIVE LIST’ OF WORK ACTIVITIES PROPOSED FOR INCORPORATION INTO THE REVISED EUROPEAN COUNCIL DIRECTIVE [8]

NORM industry	Code assigned for purposes of this paper
Extraction of rare earths from monazite	EC 1
Production of thorium compounds and manufacture of thorium-containing products	EC 2
Processing of niobium/tantalum ore	EC 3
Oil and gas production	EC 4
TiO_2 pigment production	EC 5
Thermal phosphorus production	EC 6
Zircon and zirconia industry	EC 7
Production of phosphate fertilizers	EC 8
Cement production, maintenance of clinker ovens	EC 9
Coal fired power plants, maintenance of boilers	EC 10
Phosphoric acid production	EC 11
Primary iron production	EC 12
Tin/lead/copper smelting	EC 13
Groundwater treatment	EC 14

The existence of the proposed European Commission ‘positive list’ may eventually result in an obligation to evaluate the radiation risk in the companies concerned. Such an approach could lead to the application of requirements and regulations similar to those developed for controlling the risk caused by artificial sources. However, this needs to be done in a way that takes into account the specificities of NORM industries as compared to practices involving exposure to artificial sources. For example, present exemption or clearance levels usually used in the regulation of practices are not directly applicable to NORM management. The amounts of NORM or TENORM waste are so large that treating them in the same way as radioactive waste from practices involving exposure to artificial sources cannot be done at a reasonable cost. Rather, international consensus supports a graded approach to the regulation of NORM industries. A way to implement this in practice could be to take into account the existing requirements for the assessment of occupational hazards and environmental risks in order to define the forms of regulatory control with respect to radiation protection in NORM industries. In practice, ISO management systems, environmental risk assessments and the ‘best available technology’ approach should take into consideration enhanced natural radioactivity in the industries of concern.

4. THE EUROPEAN WASTE CATALOGUE AS A REGULATORY TOOL

The solution based on the incorporation of the ‘positive list’ into national regulations is a step forward, but it does not yet provide the end user with clues on how to cope with the problem. The treatment of NORM waste or residues is a major concern in terms of the possible radiological impact on the public, because it may be disposed of at landfill sites or recycled as building material or for other applications. Identifying clearly the NORM waste of concern is a necessary step in the evaluation of the radiation risk. The European Waste Catalogue (EWC) seems to be a good basis to gather together all information about the waste, including its properties important from a radiation protection point of view. Such an idea is additionally supported by the fact that NORM waste is usually considered as a ‘common’ industrial waste rather than a nuclear or radioactive waste. Also, the consequences of NORM occurrence can be amplified by the simultaneous presence of other pollutants. Using the EWC in the regulation of NORM waste can create a bridge between radiological and non-radiological regulation. It can also be a driving force to coerce the industries of concern into embarking on the process of radiation risk monitoring and assessment.

The EWC was produced as a result of a European Commission decision[1] and is a fundamental part of the waste disposal process. It classifies both hazardous and non-hazardous waste produced pursuant to European Council Directive 75/442/EEC of 15 July 1975 on waste, and categorizes them according to what they are and how they were produced. The catalogue defines standardized nomenclatures and monitoring levels of the various waste types. The EWC codes are valid throughout the European Union and contain information on just about any waste conceivable. The EWC defines the basic necessity of monitoring certain wastes, but the monitoring level can be adapted to special needs because of requirements imposed by authorities, customers or internal regulations.

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[1] The EWC was originally established by Commission Decision 94/3/EC. It was replaced by 2000/532/EC and amended by Decisions 2001/118/EC, 2001/119/EC and 2001/216/EC. The full EWC is available from the Commission’s web-site at: http/europa.eu.int/eur-lex/en/lif/reg/en_register_15103030.html.

An analysis of the EWC in relation to NORM data available in the literature has shown that many kinds of waste listed in the EWC can be treated as at least being suspected of containing enhanced levels of natural radioactivity. The classification has been done in two institutions, independently: the Central Mining Institute (GIG), Katowice, Poland and the Federal Agency for Nuclear Control (FANC), Brussels, Belgium[2].

Wastes have been classified in accordance with the following coding system:

(a) Classification as a particular group of NORM industries according to the lists given in Tables 1 and 2 (codes IAEA 1–11 and EC 1–14);
(b) Occurrence in industrial processes involving a significant initial mass reduction, mainly thermal processes (code T1);
(c) Classification as waste created during the treatment of potable, industrial and underground water (code W1);
(d) Classification as waste created during the cleaning or treatment of atmospheric air, exhaust gases or natural gas (code A1);
(e) Classification as a particular group in accordance with data collected by the authors of this paper (code GIG 1–5).

Although there is no direct evidence, existing data from potable water treatment suggest that whenever fresh water is treated for any purpose there is likely to be quite a high risk that the associated sludge might contain enhanced concentrations of radium. A similar situation exists for all kinds of dust and solid or wet wastes from exhaust gas treatment. Regardless of the kind of gas treated, the concentration of ^{210}Pb in the gas is usually high enough to cause a high concentration of lead ^{210}Pb (and consequently ^{210}Po) in each kind of waste generated. The processed material can only amplify the effect.

In EWC, some categories of waste have been divided into two groups: those containing dangerous substances and those not containing dangerous substances. If one of the groups is classified as being at risk of containing enhanced concentrations of radionuclides of natural origin, automatically the second one is classified in the same category. The reason is that natural radionuclides were not taken into account in the definition of dangerous substances in the EWC.

The Laboratory of Radiometry (GIG) has been providing services involving the measurement of radioactivity of different materials and waste for at least 20 years. This activity resulted in quite a large database containing more than

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[2] The classification done by FANC is still an ongoing process and must not yet be considered as the official position of FANC.

TABLE 3. ADDITIONAL WORK ACTIVITIES IDENTIFIED BY GIG

	Code assigned for purposes of this paper
Coal mining	GIG 1
Production and use of abrasive materials	GIG 2
Production and use of refractories	GIG 3
Processing of potassium rich minerals	GIG 4
Paper production	GIG 5

15 000 records. Based on this, additional branches of industry need to be considered as being affected by enhanced natural radioactivity. Details are given in Table 3.

The work of FANC and GIG has resulted in the identification of those wastes for which the issue of radioactivity should be considered when their disposal is planned. The results obtained are given in Table 4.

5. CONCLUSION

Among individual types of waste already classified in the EWC, those listed in Table 4 (numbering more than 200) may be at least suspected as being TENORM or NORM. Despite such a large number of waste types already listed in the EWC, it may be, in some cases, worth distinguishing a new category of individual waste, taking into consideration concentrations of radionuclides of natural origin. For example, sludges settled at the bottom of ponds in the coal mining industry have such high activity concentrations of radium that they deserve to be isolated from other waste classified as subgroup *01 01 wastes from mineral excavation.*

The approach presented in this paper does not cover all aspects of NORM occurrence. Besides waste, existing raw materials, goods and consumables associated with NORM or TENORM should be officially listed.

The implementation of a ‘positive list’ of industries as well as the identification of potential NORM waste on basis of the EWC is only a first step in the regulatory control of NORM and TENORM that should be followed by an appropriate risk assessment.

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS

EWC code	Description	Applicable code from Tables 1–3
01 01: Wastes from mineral excavation		
01 01 01	Wastes from mineral metalliferous excavation	IAEA 4
01 01 02	Wastes from mineral non-metalliferous excavation	GIG 1; IAEA 4
01 03: Wastes from physical and chemical processing of metalliferous minerals		
01 03 04	Acid-generating tailings from processing of sulphide ore	
01 03 05	Other tailings containing dangerous substances	IAEA 1–3, 9; EC 1–3, 12, 13
01 03 07	Other wastes containing dangerous substances from physical and chemical processing of metalliferous minerals	IAEA 9
01 03 08	Dusty and powdery wastes other than those mentioned in 01 03 07	IAEA 1–3, 9; EC 1–3, 12, 13
01 03 09	Red mud from alumina production other than the wastes mentioned in 01 03 07	IAEA 9
01 03 99	Wastes not otherwise specified	IAEA 1–3, 9; EC 1–3, 12, 13
01 04: Wastes from physical and chemical processingof non-metalliferous minerals		
01 04 07	Wastes containing dangerous substances from physical and chemical processing of non-metalliferous minerals	GIG 1–3
01 04 08	Waste gravel and crushed rocks other than those mentioned in 01 04 07	GIG 1–3
01 04 09	Waste sand and clays	GIG 2, 3
01 04 10	Dusty and powdery wastes other than those mentioned in 01 04 07	GIG 1–3
01 04 11	Wastes from potash and rock salt processing other than those mentioned in 01 04 07	GIG 4
01 04 12	Tailings and other wastes from washing and cleaning of minerals other than those mentioned in 01 04 07 and 01 04 11	GIG 2, 3
01 04 13	Wastes from stone cutting and sawing other than those mentioned in 01 04 07	GIG 2, 3
01 05: Drilling muds and other drilling wastes		
01 05 04	Freshwater drilling muds and wastes	IAEA 5; EC 4
01 05 05	Oil-containing drilling muds and wastes	IAEA 5; EC 4

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
01 05 06	Drilling muds and other drilling wastes containing dangerous substances	IAEA 5; EC 4
01 05 07	Barite-containing drilling muds and wastes other than those mentioned in 01 05 05 and 01 05 06	IAEA 5; EC 4
01 05 08	Chloride-containing drilling muds and wastes other than those mentioned in 01 05 05 and 01 05 06	IAEA 5; EC 4
01 05 99	Wastes not otherwise specified	IAEA 5; EC 4
03 03: Wastes from pulp, paper and cardboard production and processing		
03 03 02	Green liquor sludge (from recovery of cooking liquor)	GIG 5
03 03 09	Lime mud waste	GIG 5
03 03 11	Sludges from on-site effluent treatment other than those mentioned in 03 03 10	W1
05 01: Wastes from petroleum refining		
05 01 02	Desalter sludges	IAEA 5; EC 4
05 01 03	Tank bottom sludges	IAEA 5; EC 4
05 01 04	Acid alkyl sludges	IAEA 5; EC 4
05 01 05	Oil spills	IAEA 5; EC 4
05 01 06	Oily sludges from maintenance operations of the plant or equipment	IAEA 5; EC 4
05 01 07	Acid tars	IAEA 5; EC 4
05 01 08	Other tars	IAEA 5; EC 4
05 01 09	Sludges from on-site effluent treatment containing dangerous substances	IAEA 5; EC 4
05 01 10	Sludges from on-site effluent treatment other than those mentioned in 05 01 09	IAEA 5; EC 4
05 01 11	Wastes from cleaning of fuels with bases	IAEA 5; EC 4
05 01 12	Oil containing acids	IAEA 5; EC 4
05 01 13	Boiler feedwater sludges	IAEA 5; EC 4
05 01 14	Wastes from cooling columns	IAEA 5; EC 4
05 01 15	Spent filter clays	IAEA 5; EC 4
05 01 16	Sulphur-containing wastes from petroleum desulphurisation	IAEA 5; EC 4

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
05 01 17	Bitumen	IAEA 5; EC 4
05 01 99	Wastes not otherwise specified	IAEA 5; EC 4
05 07: Wastes from natural gas purification and transportation		
05 07 01	Wastes containing mercury	IAEA 5; EC 4
05 07 02	Wastes containing sulphur	IAEA 5; EC 4
05 07 99	Wastes not otherwise specified	IAEA 5; EC 4
06 01: Wastes from the manufacture, formulation, supply and use (MFSU) of acids		
06 01 04	Phosphoric and phosphorous acid	IAEA 7; EC 11
06 03: Wastes from the MFSU of salts and their solutions and metallic oxides		
06 03 13	Solid salts and solutions containing heavy metals	GIG 4
06 03 15	Metallic oxides containing heavy metals	IAEA 1–3, 9; EC 1–3, 12, 13
06 03 16	Metallic oxides other than those mentioned in 06 03 15	IAEA 1–3, 9; EC 1–3, 12, 13
06 05: Sludges from on-site effluent treatment		
06 05 02	Sludges from on-site effluent treatment containing dangerous substances	IAEA 7; EC 1, 8; W1
06 05 03	Sludges from on-site effluent treatment other than those mentioned in 06 05 02	IAEA 7; EC 1, 8; W1
06 07: Wastes from the MFSU of halogens and halogen chemical processes		
06 07 03	Barium sulphate sludge containing mercury	GIG 1; EC 4; IAEA 5
06 09: Wastes from the MSFU of phosphorous chemicals and phosphorous chemical processes		
06 09 02	Phosphorous slag	EC 6
06 09 03	Calcium-based reaction wastes containing or contaminated with dangerous substances	EC 6
06 09 04	Calcium-based reaction wastes other than those mentioned in 06 09 03	EC 6
06 09 99	Wastes not otherwise specified	EC 6

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
06 11: Wastes from the manufacture of inorganic pigments and opacifiers		
06 11 01	Calcium-based reaction wastes from titanium dioxide production	IAEA 6; EC 5
06 13: Wastes from inorganic chemical processes not otherwise specified		
06 13 02	Spent activated carbon (except 06 07 02)	W1, A1
06 13 03	Carbon black	A1
06 13 05	Soot	A1
08 02: Wastes from MFSU of other coatings, including ceramic materials		
08 02 02	Aqueous sludges containing ceramic materials	GIG 2
08 02 03	Aqueous suspensions containing ceramic materials	GIG 2
10 01: Wastes from power stations and other combustion plants, except group 19		
10 01 01	Bottom ash, slag and boiler dust (excluding boiler dust mentioned in 10 01 04)	IAEA 10; EC 10; T1
10 01 02	Coal fly ash	IAEA 10; EC 10; T1
10 01 03	Flyash from peat and untreated wood	T1
10 01 04	Oil fly ash and boiler dust	T1
10 01 05	Calcium-based reaction wastes from flue-gas desulphurization in solid form	T1, A1
10 01 07	Calcium-based reaction wastes from flue-gas desulphurization in sludge form	T1, A1
10 01 13	Fly ash from emulsified hydrocarbons used as fuel	T1
10 01 14	Bottom ash, slag and boiler dust from co-incineration containing dangerous substances	T1
10 01 15	Bottom ash, slag and boiler dust from co-incineration other than those mentioned in 10 01 14	T1
10 01 16	Fly ash from co-incineration containing dangerous substances	T1
10 01 17	Fly ash from co-incineration other than those mentioned in 10 01 16	T1
10 01 18	Wastes from gas cleaning containing dangerous substances	A1
10 01 19	Wastes from gas cleaning other than those mentioned in 10 01 05, 10 01 07 and 10 01 18	A1

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 01 20	Sludges from on-site effluent treatment containing dangerous substances	W1
10 01 21	Sludges from on-site effluent treatment other than those mentioned in 10 01 20	W1
10 01 22	Aqueous sludges from boiler cleaning containing dangerous substances	T1
10 01 23	Aqueous sludges from boiler cleansing other than those mentioned in 10 01 22	T1
10 01 24	Sands from fluidized beds	T1
10 01 26	Wastes from cooling-water treatment	W1
10 02: Wastes from the iron and steel industry		
10 02 02	Unprocessed slag	T1
10 02 07	Solid wastes from gas treatment containing dangerous substances	IAEA 9; EC 12
10 02 08	Solid wastes from gas treatment other than those mentioned in 10 02 07	A1
10 02 10	Mill scales	GIG 3
10 02 12	Wastes from cooling-water treatment other than those mentioned in 10 02 11	W1
10 02 13	Sludges and filter cakes from gas treatment containing dangerous substances	A1
10 02 14	Sludges and filter cakes from gas treatment other than those mentioned in 10 02 13	A1
10 02 15	Other sludges and filter cakes	A1; W1
10 03: Wastes from aluminium thermal metallurgy		
10 03 04	Primary production slags	T1
10 03 05	Waste alumina	IAEA 9
10 03 08	Salt slags from secondary production	T1
10 03 09	Black drosses from secondary production	T1
10 03 19	Flue-gas dust containing dangerous substances	IAEA 9; A1
10 03 20	Flue-gas dust other than those mentioned in 10 03 19	IAEA 9; A1
10 03 21	Other particulates and dust (including ball-mill dust) containing dangerous substances	A1

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 03 22	Other particulates and dust (including ball-mill dust) other than those mentioned in 10 03 21	A1; GIG 3
10 03 23	Solid wastes from gas treatment containing dangerous substances	A1
10 03 24	Solid wastes from gas treatment other than those mentioned in 10 03 23	A1
10 03 25	Sludges and filter cakes from gas treatment containing dangerous substances	A1; W1
10 03 26	Sludges and filter cakes from gas treatment other than those mentioned in 10 03 25	A1; W1
10 03 27	Wastes from cooling-water treatment containing oil	W1
10 03 28	Wastes from cooling-water treatment other than those mentioned in 10 03 27	W1
10 04: Wastes from lead thermal metallurgy		
10 04 01	Slags from primary and secondary production	IAEA 9; EC 13
10 04 04	Flue-gas dust	IAEA 9; EC 13
10 04 05	Other particulates and dust	A1
10 04 06	Solid wastes from gas treatment	A1
10 03 29	Wastes from treatment of salt slags and black drosses containing dangerous substances	T1
10 03 30	Wastes from treatment of salt slags and black drosses other than those mentioned in 10 03 29	T1
10 04 07	Sludges and filter cakes from gas treatment	A1; W1
10 04 09	Wastes from cooling-water treatment containing oil	W1
10 04 10	Wastes from cooling-water treatment other than those mentioned in 10 04 09	W1
10 05: Wastes from zinc thermal metallurgy		
10 05 01	Slags from primary and secondary production	IAEA 9
10 05 03	Flue-gas dust	IAEA 9
10 05 04	Other particulates and dust	A1
10 05 05	Solid waste from gas treatment	A1
10 05 06	Sludges and filter cakes from gas treatment	A1; W1

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 05 08	Wastes from cooling-water treatment containing oil	W1
10 05 09	Wastes from cooling-water treatment other than those mentioned in 10 05 08	W1
10 06: Wastes from copper thermal metallurgy		
10 06 01	Slags from primary and secondary production	IAEA 9; EC 13, T1
10 06 02	Dross and skimmings from primary and secondary production	T1
10 06 03	Flue-gas dust	A1; IAEA 9; EC 13,
10 06 04	Other particulates and dust	A1
10 06 06	Solid wastes from gas treatment	A1
10 06 07	Sludges and filter cakes from gas treatment	A1; W1
10 06 09	Wastes from cooling-water treatment containing oil	W1
10 06 10	Wastes from cooling-water treatment other than those mentioned in 10 06 09	W1
10 07: Wastes from silver, gold and platinum thermal metallurgy		
10 07 01	Slags from primary and secondary production	T1
10 07 02	Dross and skimmings from primary and secondary production	T1
10 07 03	Solid wastes from gas treatment	A1
10 07 04	Other particulates and dust	A1
10 07 05	Sludges and filter cakes from gas treatment	A1
10 07 07	Wastes from cooling-water treatment containing oil	W1
10 07 08	Wastes from cooling-water treatment other than those mentioned in 10 07 07	W1
10 08: Wastes from other non-ferrous thermal metallurgy		
10 08 04	Particulates and dust	IAEA 2, 3, 9; EC 2, 3, 13
10 08 08	Salt slag from primary and secondary production	IAEA 2, 3, 9; EC 2, 3, 13
10 08 09	Other slags	IAEA 2, 3, 9; EC 2, 3, 13

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 08 10	Dross and skimmings that are flammable or emit, upon contact with water, flammable gases in dangerous quantities	T1
10 08 11	Dross and skimmings other than those mentioned in 10 08 10	T1
10 08 15	Flue-gas dust containing dangerous substances	IAEA 2, 3, 9; EC 2, 3, 13; A1
10 08 16	Flue-gas dust other than those mentioned in 10 08 15	IAEA 2, 3, 9; EC 2, 3, 13; A1
10 08 17	Sludges and filter cakes from flue-gas treatment containing dangerous substances	A1
10 08 18	Sludges and filter cakes from flue-gas treatment other than those mentioned in 10 08 17	A1
10 08 19	Wastes from cooling-water treatment containing oil	W1
10 08 20	Wastes from cooling-water treatment other than those mentioned in 10 08 19	W1
10 09: Wastes from casting of ferrous pieces		
10 09 03	Furnace slag	GIG 3
10 09 05	Casting cores and moulds which have not undergone pouring containing dangerous substances	GIG 3
10 09 06	Casting cores and moulds which have not undergone pouring other than those mentioned in 10 09 05	GIG 3
10 09 07	Casting cores and moulds which have undergone pouring containing dangerous substances	GIG 3
10 09 08	Casting cores and moulds which have undergone pouring other than those mentioned in 10 09 07	GIG 3
10 09 09	Flue-gas dust containing dangerous substances	A1
10 09 10	Flue-gas dust other than those mentioned in 10 09 09	A1
10 09 11	Other particulates containing dangerous substances	T1
10 09 12	Other particulates other than those mentioned in 10 09 11	T1
10 10: Wastes from casting of non-ferrous pieces		
10 10 05	Casting cores and moulds which have not undergone pouring, containing dangerous substances	IAEA 8; EC 7
10 10 06	Casting cores and moulds which have not undergone pouring, other than those mentioned in 10 10 05	IAEA 8; EC 7

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 10 07	Casting cores and moulds which have undergone pouring, containing dangerous substances	IAEA 8; EC 7
10 10 08	Casting cores and moulds which have undergone pouring, other than those mentioned in 10 10 07	IAEA 8; EC 7
10 10 09	Flue-gas dust containing dangerous substances	A1
10 10 10	Flue-gas dust other than those mentioned in 10 10 09	A1
10 10 11	Other particulates containing dangerous substances	T1
10 10 12	Other particulates other than those mentioned in 10 10 11	T1
10 11: Wastes from manufacture of glass and glass products		
10 11 05	Particulates and dust	A1
10 11 13	Glass-polishing and -grinding sludge containing dangerous substances	GIG 2
10 11 14	Glass-polishing and -grinding sludge other than those mentioned in 10 11 13	GIG 2
10 11 15	Solid wastes from flue-gas treatment containing dangerous substances	GIG 2
10 11 16	Solid wastes from flue-gas treatment other than those mentioned in 10 11 15	A1
10 11 17	Sludges and filter cakes from flue-gas treatment containing dangerous substances	A1
10 11 18	Sludges and filter cakes from flue-gas treatment other than those mentioned in 10 11 17	A1
10 11 19	Solid wastes from on-site effluent treatment containing dangerous substances	W1
10 11 20	Solid wastes from on-site effluent treatment other than those mentioned in 10 11 19	W1
10 12: Wastes from manufacture of ceramic goods, bricks, tiles and construction products		
10 12 01	Waste preparation mixture before thermal processing	IAEA 8; EC 7
10 12 03	Particulates and dust	A1
10 12 05	Sludges and filter cakes from gas treatment	A1
10 12 06	Discarded moulds	GIG 3
10 12 08	Waste ceramics, bricks, tiles and construction products (after thermal processing)	IAEA 8; EC 7

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
10 12 09	Solid wastes from gas treatment containing dangerous substances	A1
10 12 10	Solid wastes from gas treatment other than those mentioned in 10 12 09	A1
10 12 11	Wastes from glazing containing heavy metals	T1; IAEA 8; EC 7
10 12 12	Wastes from glazing other than those mentioned in 10 12 11	T1
10 12 13	Sludge from on-site effluent treatment	W1
10 13: Wastes from manufacture of cement, lime and plaster and articles and products made from them		
10 13 06	Particulates and dust (except 10 13 12 and 10 13 13)	A1
10 13 07	Sludges and filter cakes from gas treatment	A1
10 13 09	Wastes from asbestos-cement manufacture containing asbestos	EC 9
10 13 12	Solid wastes from gas treatment containing dangerous substances	A1; EC 9
10 13 13	Solid wastes from gas treatment other than those mentioned in 10 13 12	A1; EC 9
10 14: Waste from crematoria		
10 14 01	Waste from gas cleaning containing mercury	A1
12 01: Wastes from shaping and physical and mechanical surface treatment of metals and plastics		
12 01 13	Welding wastes	IAEA 2; EC 2
12 01 14	Machining sludges containing dangerous substances	GIG 2
12 01 15	Machining sludges other than those mentioned in 12 01 14	GIG 2
12 01 16	Waste blasting material containing dangerous substances	A1
12 01 17	Waste blasting material other than those mentioned in 12 01 16	A1
12 01 18	Metal sludge (grinding, honing and lapping sludge) containing oil	GIG 2
12 01 19	Readily biodegradable machining oil	
12 01 20	Spent grinding bodies and grinding materials containing dangerous substances	GIG 2
12 01 21	Spent grinding bodies and grinding materials other than those mentioned in 12 01 20	GIG 2

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
16 08: Spent catalysts		
16 08 01	Spent catalysts containing gold, silver, rhenium, rhodium, palladium, iridium or platinum (except 16 08 07)	IAEA 5; EC 4
16 08 02	Spent catalysts containing dangerous transition metals (3) or dangerous transition metal compounds	IAEA 5; EC 4
16 08 03	Spent catalysts containing transition metals or transition metal compounds not otherwise specified	IAEA 5; EC 4
16 08 04	Spent fluid catalytic cracking catalysts (except 16 08 07)	IAEA 5; EC 4
16 08 05	Spent catalysts containing phosphoric acid	IAEA 5; EC 4
16 08 06	Spent liquids used as catalysts	IAEA 5; EC 4
16 08 07	Spent catalysts contaminated with dangerous substances	IAEA 5; EC 4
16 11: Waste linings and refractories		
16 11 01	Carbon-based linings and refractories from metallurgical processes containing dangerous substances	IAEA 8; EC 7, 9
16 11 02	Carbon-based linings and refractories from metallurgical processes others than those mentioned in 16 11 01	IAEA 8; EC 7, 9; GIG 3
16 11 03	Other linings and refractories from metallurgical processes containing dangerous substances	IAEA 8; EC 7, 9; GIG 3
16 11 04	Other linings and refractories from metallurgical processes other than those mentioned in 16 11 03	IAEA 8; EC 7, 9; GIG 3
16 11 05	Linings and refractories from non-metallurgical processes containing dangerous substances	IAEA 8; EC 7, 9; GIG 3
16 11 06	Linings and refractories from non-metallurgical processes others than those mentioned in 16 11 05	IAEA 8; EC 7, 9; GIG 3
17 01: Concrete, bricks, tiles and ceramics		
17 01 03	Tiles and ceramics	IAEA 8; EC 7
17 01 06	Mixtures of, or separate fractions of concrete, bricks, tiles and ceramics containing dangerous substances	IAEA 8; EC 7
17 01 07	Mixtures of concrete, bricks, tiles and ceramics other than those mentioned in 17 01 06	IAEA 8; EC 7
17 04: Metals (including their alloys)		
17 04 05	Iron and steel	GIG 1; IAEA 7, 5; EC 4

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
17 04 06	Tin	GIG 1; IAEA 7, 5; EC 4
17 04 07	Mixed metals	GIG 1; IAEA 7, 5; EC 4
17 04 09	Metal waste contaminated with dangerous substances	GIG 1; IAEA 7, 5; EC 4
19 01: Wastes from incineration or pyrolysis of waste		
19 01 05	Filter cake from gas treatment	A1
19 01 06	Aqueous liquid wastes from gas treatment and other aqueous liquid wastes	W1
19 01 07	Solid wastes from gas treatment	A1
19 01 10	Spent activated carbon from flue-gas treatment	A1
19 01 11	Bottom ash and slag containing dangerous substances	T1
19 01 12	Bottom ash and slag other than those mentioned in 19 01 11	T1
19 01 13	Fly ash containing dangerous substances	T1
19 01 14	Fly ash other than those mentioned in 19 01 13	T1
19 01 15	Boiler dust containing dangerous substances	T1; GIG 3
19 01 16	Boiler dust other than those mentioned in 19 01 15	T1; GIG 3
19 01 17	Pyrolysis wastes containing dangerous substances	T1
19 01 18	Pyrolysis wastes other than those mentioned in 19 01 17	T1
19 01 19	Sands from fluidized beds	T1
19 09: Wastes from the preparation of water intended for human consumption or water for industrial use		
19 09 01	Solid waste from primary filtration and screenings	IAEA 11; EC 14; W1
19 09 02	Sludges from water clarification	IAEA 11; EC 14; W1
19 09 03	Sludges from decarbonation	IAEA 11; EC 14; W1
19 09 04	Spent activated carbon	IAEA 11; EC 14; W1
19 09 05	Saturated or spent ion exchange resins	IAEA 11; EC 14; W1

TABLE 4. EWC WASTE TYPES FOR WHICH DISPOSAL MAY INVOLVE RADIOACTIVITY CONSIDERATIONS (cont.)

EWC code	Description	Applicable code from Tables 1–3
19 09 06	Solutions and sludges from regeneration of ion exchangers	IAEA 11; EC 14; W1
19 09 99	Wastes not otherwise specified	IAEA 11; EC 14; W1
19 10: Wastes from shredding of metal-containing wastes		
19 10 01	Iron and steel waste	IAEA 5, 7; EC 4, 11; GIG 1
19 10 03	Fluff-light fraction and dust containing dangerous substances	A1
19 10 04	Fluff-light fraction and dust other than those mentioned in 19 10 03	A1
19 13: Wastes from soil and groundwater remediation		
19 13 05	Sludges from groundwater remediation containing dangerous substances	GIG 1
19 13 06	Sludges from groundwater remediation other than those mentioned in 19 13 05	GIG 1
19 13 07	Aqueous liquid wastes and aqueous concentrates from groundwater remediation containing dangerous substances	GIG 1
19 13 08	Aqueous liquid wastes and aqueous concentrates from groundwater remediation other than those mentioned in 19 13 07	GIG 1

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TOWARDS AN EVIDENCE BASED SCORE CARD FOR ALIGNING RISK MANAGEMENT AND SUSTAINABILITY GOALS FOR ESSENTIAL NORM INDUSTRIES: CASE STUDY — PHOSPHATES

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Abstract

Approaches to regulating NORM industries risk suffering blight from over-conservative methodologies, whether based on worst case models, extreme event scenarios or unmediated application of the precautionary principle: the outcome can be a significant overestimation of risk and a consequent penalty on both producers and consumers in terms of access to and affordability of the intermediate and end products those industries provide. In particular, for historical reasons derived perhaps from the potentially distracting regulatory focus on what is usually trace radioactivity in products and by-products containing NORM, there is a damaging tendency to seek risk management models and best practices from nuclear industries in general rather than from those sectors to which the end products of NORM industries are specifically aligned. This risk is particularly visible in the phosphate sector, an industry now pivotal to long term security and sustainability in both food production and energy supply, plant based or nuclear. Premised on a companion paper which sets out the theory of ‘constructive regulation’, presented in 2008 at the 12th International Congress of the International Radiation Protection Association, Buenos Aires, this paper proposes the use of an evidence based score carding system to ensure the future alignment of risk management and sustainability goals for NORM industries, starting with phosphates. The score card elements are broken out into three primary categories along the lines defined in the concept of triple bottom line performance measurement, comprising economic, social and environmental elements. The question is put as to what role constructive regulation and best practices can play in ensuring that the outcome of the regulatory process is the preservation and enhancement of the capability of these industries to deliver sustainable returns to the customers and stakeholders who depend on them. Score carding will facilitate transparent, objective decision making and effective performance monitoring in both the short and long term, as measured against triple bottom line expectations.

1. INTRODUCTION: CONSTRUCTIVE REGULATION OF NORM INDUSTRIES

The regulation of NORM industries is in its relative infancy. A number of key challenges face the NORM regulatory body at national and international levels as the regulatory process matures. These include:

(a) Developing an adequate and coherent definition of a ‘NORM industry’ to which a given industry or process can reasonably be assigned;
(b) Creating coherent and consistent standards from which a regulatory method can be developed;
(c) Using evidence and scientific practice as the means of measuring and regulating real hazards in processes and outcomes;
(d) Reducing the dependence on questionable risk assessments, derived from hyper-conservative, ‘worst case’ models or implausible scenarios as a basis for regulation.

The long term goal of the regulatory process is not in doubt — the achievement of safe, sustainable processes, creating safe, beneficial products yielding positive returns to customers, shareholders and stakeholders at no risk, or at acceptable risk, to occupational, public and environmental health and safety. How this goal is best reached is open for discussion. The concept of ‘constructive regulation’ [1] was first proposed before the financial crisis of 2008–2009 and the emergence of the new economic order, focused on G20 countries. It seems to have gained in relevance from those recent changes because the status of policy objectives such as resource conservation and sustainability has changed in the wake of global recession from ‘nice to have’ to ‘must have’. The impact on regulatory practice is most evident in attitudes to and purposes for ‘waste’, where it is no longer acceptable to define something as a waste without further recourse. If conservation is the goal, a ‘waste’ can only be called such when no foreseen or foreseeable use for that material is apparent. One consequence of this change is that the regulatory body will need both to justify any initial restrictions or conditions placed on NORM industries on the basis of measurable risk or hazard; a second is to continue to justify such measures on the basis of delivered benefit to direct and indirect stakeholders. This paper proposes a way to assist the regulatory body in meeting this objective, while also rendering the process of regulation constructive — meaning transparent and accountable — by the use of a balanced score card. The score card proposed has its roots in performance management, but is adapted specifically for NORM.

1.1. Score cards and dashboards

The balanced score card is a strategic planning and management device widely used in industry and commerce. The objectives are to:

(a) Align operational or business activities to the vision and strategy of the organization;
(b) Improve internal and external communications;
(c) Monitor organizational performance against strategic goals [2].

While the phrase balanced score card was coined in the early 1990s [3, 4], the roots of this approach to process and outcome measurement are deep. They include the pioneering work of General Electric on performance measurement reporting in the 1950s and, yet earlier, the work of French process engineers in the 1930s who created the Tableau de Bord — literally, a ‘dashboard’ for monitoring industrial processes. [5]

Because of its ability to capture and arrange feedback, the balanced score card has evolved from its early use as a simple performance measurement framework to a full strategic planning and management system. The score card device translates an organization’s strategic plan from passive to active status, reaching down into the ‘marching orders’ for the organization on a daily basis. This is particularly helpful in the communication process with external agents, notably shareholders and stakeholders, since it not only provides a narrative of what the organization is doing at any one time, but why it is doing it and what the anticipated benefits are of so doing. In this way it enables an organization also to reach a new point of equilibrium in its operational practices, one which meets the Nash criteria for success in developing a negotiated, cooperative solution (win–win) to an apparently intractable problem [6], which is one of the success conditions for achieving constructive regulation.

At the heart of the score card are the organizational vision and strategy — what are we tasked to do and how are we going to do it? Success depends on aligning the ‘what and why’ with the ‘how’. This alignment process is managed by four key indicators:

(1) Customer relations and feedback;
(2) Business processes;
(3) Learning and growth;
(4) Financial performance.

The key differentiator of score carding from other performance measurement techniques is that it adds strategic non-financial performance measures to traditional financial metrics to give managers and executives a better contextualised view both of performance and of outcomes, especially in knowledge intensive businesses. In the wake of the 1990s internet revolution, financial measures in isolation were deemed inadequate for guiding and evaluating the continuous journey that knowledge based companies would have to take to create future value. Sustainability would be best achieved through continuous investment in customers, suppliers, employees, processes and technological innovation. While such businesses recognized the need for a more holistic, multi-dimensional approach to operational performance, not all regulatory bodies followed suit. They, by contrast, focused entirely on the 'customer' part of the system, to which they applied a one-dimensional model derived from pathway analysis. The underlying supposition was, in effect, that risk was inherent (therefore identical with hazard) and that the only defensible position for a state agency to take on risk was effectively zero tolerance. If, as for NORM, the aetiology of that risk is naturally occurring, then the only behavioural approach that could be supported was quantitative reduction (lowering the threshold) even when (a) there was no evidence that the inherent hazard actually did cause harm and (b) no evidence over time that the reduced threshold translated into increased individual or societal benefit from a public health point of view. A consequence, for example, for phosphate regulation was that it emphasized the prevention of pollution (phosphate as prejudicial input) rather than the opportunity to extract or recover phosphate from waste (phosphate as valuable resource).

In the USA, the zero tolerance approach forced a major negative externality onto the community in the form of compulsory land stacking phosphogypsum with low concentrations of NORM. The approach also ignored the negative environmental impact of the chosen containment method [7]. Had a balanced score carding approach been adopted, it would probably have demonstrated that the use of phosphogypsum would deliver a higher social return on investment than containment, while regulatory objectives would better have focused on managing perceived and actual risk through better public awareness, enhancing production processes and better educating and training the workforce.

The continued reliance on the part of some regulatory bodies on a single end-point model of risk is all the more problematic as the new economic order realigns its objectives to resource conservation and sustainability. These policies deliver benefit both through business process innovation and investment in learning and growth channels. If these are ignored or blocked by the regulatory body, there is a fundamental conflict between two policy objectives, as the policy of absolute safety collides with the policy of sustainability. Sustainability can only be achieved if safety is understood to have both absolute and relative characteristics. The compelling reason to adopt score carding is that it optimizes absolute and relative safety policies in concert rather than placing them in opposition. So what and how should the score card score?

1.2. The triple bottom line model

The triple bottom line (TBL) method of parsing and presenting enterprise performance has made rapid progress since its formulation in 1996 [8]. Against a similar background of changes in global business culture that gave birth to the balanced score card, TBL recognizes that financial measures alone do not adequately capture the performance of a given enterprise or organization to its stockholders and other stakeholders, nor even act as reliable predictors of its future longevity. TBL argues that in addition to financial or economic performance, an enterprise must also show benefit in terms of social return (social capital, social return on investment, public good) and environmental return (minimized negative legacy, low ‘footprint’). Taking a score card approach to TBL allows an organization to roll up its environmental performance in terms of pollution and environmental disturbance in general, but also to drill down into particular indicators such as GHG–CO_2 emissions or monitor use of specific inputs such as energy and water.

Examples such as nuclear power generation or commercial use of genetically modified crops illustrate what happens when stakeholders perceptions of risk are ignored. In both instances, when the issues of social and environmental return were left unaddressed, public (stakeholder) resistance stopped successful industries dead in their tracks. The benefit of appraisal by score carding is self-interested, forcing stakeholder concerns and expectations onto the performance ‘balance sheet’. If the phosphate fertilizer industry is to avoid similar trouble, it may find the combination of TBL analysis and sustainable score carding a good basis both for aligning its internal objectives with those of society as a whole, while also building an effective communication platform with stakeholder in the process.

As shown in Fig. 1, the score card methodology blends very simply with the TBL approach and sustainability measures can be derived from the score card with little further modification.

In Fig. 1, the traditional score card is presented in the upper half of the content boxes and the sustainability indicators are overlaid in the lower half. The dependency on information and knowledge, which is at the heart of the original score card concept works, if anything, even more convincingly for the TBL approach. Under TBL the need for specialized knowledge and expertise grows, but also diversifies. Business processes refocus from a commoditized (maximized profit) outcome, to an outcome in which conservation and reuse are a part of the concept of ‘profit’ — at least in the accounting technical sense that on

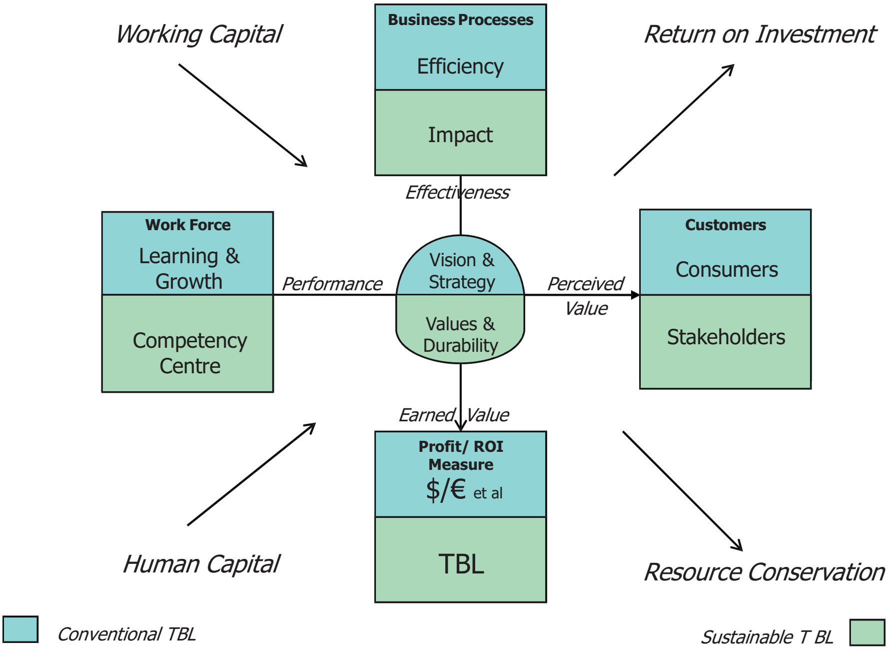


FIG. 1. Sustainability — an adapted balanced score card.

the enterprise balance sheet retained earnings is equivalent to retained resources. This has the further consequence of overlaying additional obligations on the four components of the score card, so that:

(a) Learning and growth now embrace a formalized competency centre requirement with certification, meaning that training and capacity building are ‘must dos’ not ‘nice to haves’;
(b) Efficiency in business processes is moderated by the expected impact those processes may have on stakeholders and wider society (as for example in banking);
(c) The role of the consumer is balanced by the need to satisfy both direct and indirect stakeholders;
(d) TBL is taken as the metric for measuring financial performance, rather than simply net cash or net present value.

1.3. Risk and hazard

The focus of any negotiated, cooperative solution to optimizing the balance of absolute and relative risk in the Nash model is on risk not on hazard. In the

Nash paradigm, if one starts from hazard, the outcome is de facto non-cooperative. In that Nash is able to demonstrate mathematically that there are certain conflicted situations which can only be resolved by both parties agreeing a cooperative solution, from which both win, and without which both lose, so the theorem can also demonstrate that the extrapolation of a modelled risk into a perceived hazard actually creates the conditions for a ‘non-cooperative’ game in which both parties have to lose. This is the situation caused by the PG Rule of the USEPA [9]. Using a combination of a questionable risk assessment model, based on an implausible scenario and combining it with definitional assertion that PG is a hazardous waste, the Rule first elides an estimate of risk into an absolute hazard, then uses the defined hazard as an absolute to justify mandatory stacking of PG. Thus by definitional rather than scientific means, USEPA removes from the table any prospect of a cooperative solution, even if one were warranted evidentially. This example is all the more significant in that it points to a high level of ‘risk’ for NORM industries in general that the same elision may occur in the regulatory process, from a mathematical hypothesis into a reified hazard, which then becomes a de facto barrier.

Risk management, in the end, is a contingent not an absolute process, an assessment of probability, not certainty, of outcome. The problem with a number of modelled and scenario based ‘risk’ assessments is that they effectively conflate risk (a hypothesis) with hazard (an absolute), and then derive a proposed practice or regulation from the declared hazard rather than an evidentially warranted one.

2. PHOSPHATES — AN ESSENTIAL NORM INDUSTRY

There is perhaps no substance more central to the realization of the policy of sustainability than phosphorus. Essential to all life forms, with no substitutes, phosphorus reserves are being depleted at an increasing pace at a time when concern is growing as to just how long reserves will last [10–12]. It is not the purpose of this paper to contribute to the depletion debate; but it is of concern if poor regulatory practices contribute to inefficient operational processes, including resource recovery, to compound the depletion problem, or worse if concerns about radionuclides and heavy metals stop the industry in its tracks.

2.1. A new sustainable business model

What is at issue is moving the thought process along in respect to the way phosphate as an essential industry can respond to the challenge of sustainability, while receiving a ‘constructive’ level of support from the regulatory body as to how to make the necessary adjustments in both financial and operational business

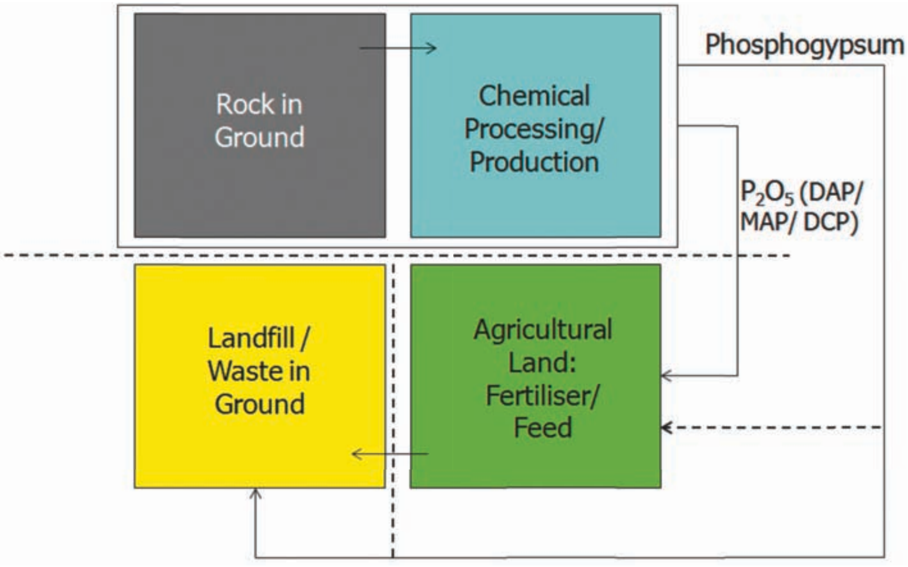


FIG. 2. The Commoditized phosphate value chain.

models. The current model is essentially commodity driven. Di-ammonium phosphate (DAP), mono-ammonium phosphate and di-calcium phosphate represent the great bulk of the world market in fertilizers and animal feed supplements. The current value chain is very short in nature consisting essentially of four steps:

(1) Mining the rock out of the ground, with associated beneficiation;
(2) Chemical processing to extract P (reported as P_2O_5) in phosphoric acid;
(3) Formulation and granulation as a fertilizer;
(4) Application to the soil or inclusion as a feed supplement.

At that point the value chain stops. As is shown in Fig. 2, while some PG finds use in soils as an amendment, much is simply stacked, a practice required by law in some countries. So ends the value chain and in many cases the stack itself turns into a liability, changing status from resource to waste.

Applying the principles of resource conservation and sustainability, but equally the objective of maximizing return on investment from phosphate rock mined, it is possible to construct a model of the phosphate industry, as shown in Fig. 3, that aligns it to the utility sector. Phosphate rock is extracted from the ground, processed and then reapplied, primarily to soil, but also to animals. What is added is the principle of recovery and reuse, driven by the premise that P

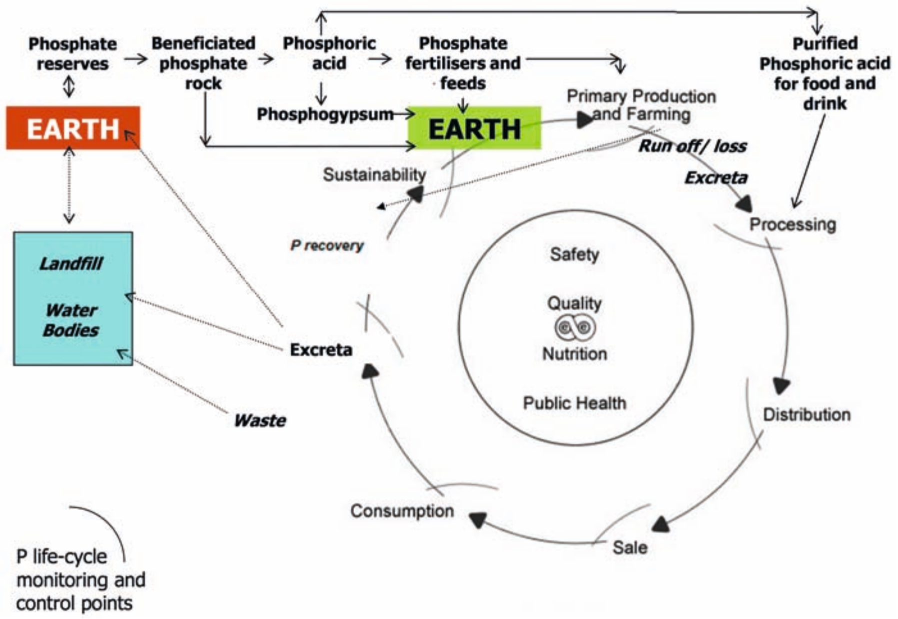


FIG. 3. The earth to earth (E2E) phosphate life-cycle model.

product streams are only wastes inasfar as we declare them to be so. The theory of conservation is thus applied under the rubric ‘earth to earth’ (E2E) [1], as shown in Fig. 3, to extend the steps in the value chain in principle indefinitely, since there is at least in theory no limit to the number of times the P can be recovered and reapplied.

Once the vicious circle of commoditization is broken, a new, sustainable model for P production and use can be constructed, as shown in Fig. 4. This has the inherent capability of moving production and consumption progressively towards equilibrium [13].

Such an approach also delves into the original process stream to seek additional value than just the P itself, with two broad implications:

(i) It is of strategic value for energy security that the U present in the phosphoric acid be removed for the nuclear fuel cycle;
(ii) It will be a pressing requirement for the future that the P industry improves the recovery rate of phosphate between the mining step and chemical processing; for while P recovery in the digestion process (attack tank) is very high, before the P gets to that point estimates suggest that 20–30% of the P is lost back into the spoil or the off-take from beneficiation [12].

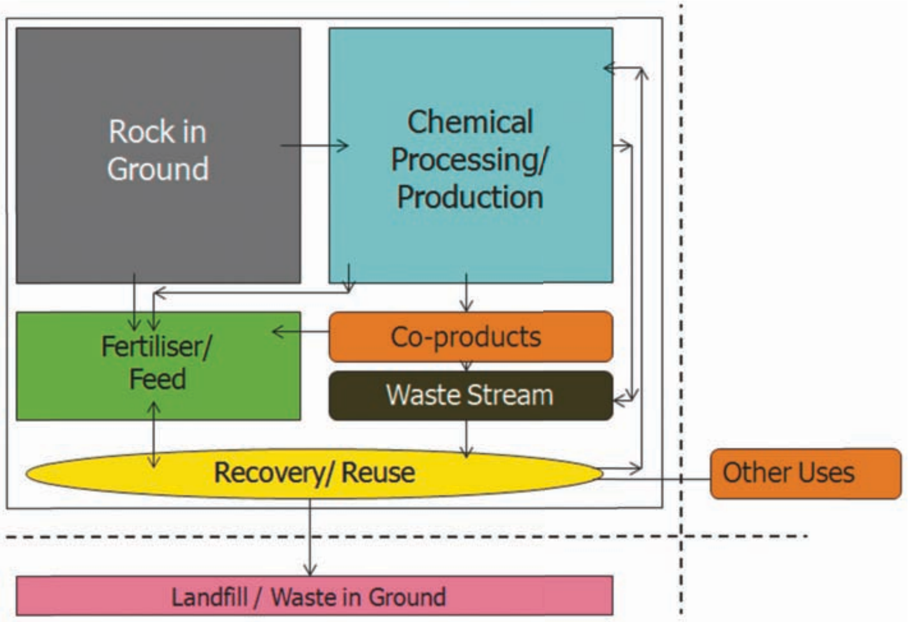


FIG.4. The 'utility' phosphate value chain.

The E2E model (Fig. 3) has the further merit of conforming to the revised definition of waste. Only that part of the product and production stream goes to landfill that cannot be recovered or stockpiled for any other use.

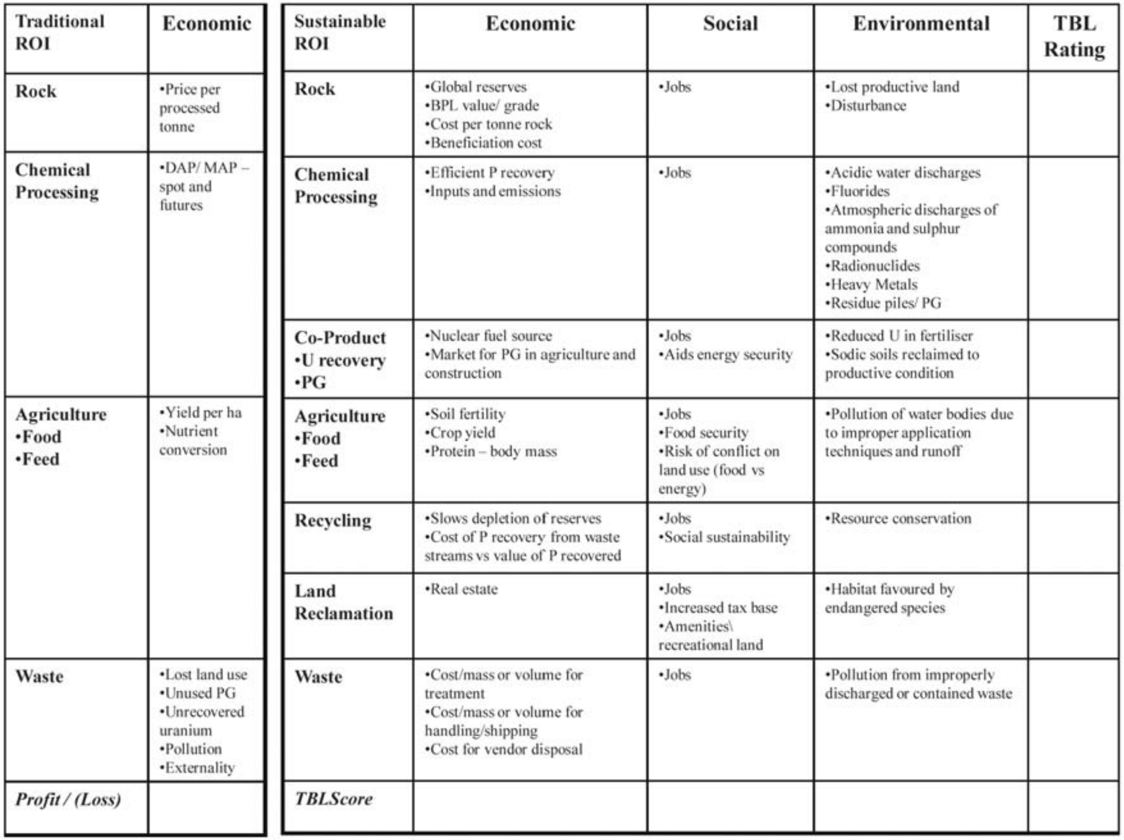
2.2. Balanced score cards and constructive regulation

In terms of constructive regulation, it is open to the policy maker, assisted by the regulatory body, to use regulatory levers, such as waste directives, to encourage this transformation of the value chain from a four step linear model to an infinitely recursive one. The regulatory body needs to put that objective into the heart of the vision and strategy on the balanced score card (see Fig. 1) and work through the implications for regulatory behaviour accordingly. To assist this process, Table 1 offers a sample score card, comparing the current commodity approach with a potential future sustainable one.

2.3. Evidence and outcomes

Evidence and outcomes are the most substantive components of a balanced score card approach to assessments of risk and hazard in any industry, including NORM. The impact of this approach can best be measured in touchstone situations

TABLE 1. COMPARATIVE BALANCED SCORE CARDS



Traditional ROI	Economic
Rock	•Price per processed tonne
Chemical Processing	•DAP/ MAP – spot and futures
Agriculture **•Food** **•Feed**	•Yield per ha •Nutrient conversion
Waste	•Lost land use •Unused PG •Unrecovered uranium •Pollution •Externality
Profit / (Loss)	

Sustainable ROI	Economic	Social	Environmental	TBL Rating
Rock	•Global reserves •BPL value/ grade •Cost per tonne rock •Beneficiation cost	•Jobs	•Lost productive land •Disturbance	
Chemical Processing	•Efficient P recovery •Inputs and emissions	•Jobs	•Acidic water discharges •Fluorides •Atmospheric discharges of ammonia and sulphur compounds •Radionuclides •Heavy Metals •Residue piles/ PG	
Co-Product **•U recovery** **•PG**	•Nuclear fuel source •Market for PG in agriculture and construction	•Jobs •Aids energy security	•Reduced U in fertiliser •Sodic soils reclaimed to productive condition	
Agriculture **•Food** **•Feed**	•Soil fertility •Crop yield •Protein – body mass	•Jobs •Food security •Risk of conflict on land use (food vs energy)	•Pollution of water bodies due to improper application techniques and runoff	
Recycling	•Slows depletion of reserves •Cost of P recovery from waste streams vs value of P recovered	•Jobs •Social sustainability	•Resource conservation	
Land Reclamation	•Real estate	•Jobs •Increased tax base •Amenities\ recreational land	•Habitat favoured by endangered species	
Waste	•Cost/mass or volume for treatment •Cost/mass or volume for handling/shipping •Cost for vendor disposal	•Jobs	•Pollution from improperly discharged or contained waste	
TBLScore				

such as the handling of phosphogypsum. In the USA in particular, the Phosphogypsum Rule [9] mandates the stacking of phosphogypsum and effectively prohibits its use through a highly onerous legal and technical permitting procedure that fits the Nash definitional criteria of a ‘non-cooperative’ game.

This approach sets evidence and outcome aside. The practice is justified by a risk calculation, based on a particular scenario. What is not done is a balanced calculation of the long term TBL outcome (impact, consequence) of creating large stacks of PG against the outcome of use, partly because use, being effectively prohibited, no longer yields any evidence base, at least in the USA, as to what long term consequences it may have.

Fortunately, in the era of globalization, there are data sets from other settings, notably Huelva, Spain, where there is 70 years of experience of using PG on soils and crops [14]. Evidence and outcomes from this setting are well documented as a result of a five year landmark study, exemplary in its use of the evidence based approach. For this paper, the length of time over which phosphogypsum has been used is the one detail worthy of further comment. Studying potentially genetically transmissible risk tends be conducted over three generations, with the likelihood high that if no abnormalities show up in that time frame, they are unlikely to be present. Such is the case in the Huelva setting.

2.4. Backward induction — from depletion to sustainable equilibrium

As a first step in validating the score card approach to improved decision making processes in the regulation of NORM industries, a brief schematized example is offered in Fig. 5. The enabling condition for this approach is the understanding that P can be regarded as being in a closed rather than an open system [15].

One way to align the objectives of food security with P resource conservation is to apply backward induction theory to the whole phosphate value stream, including both phosphogypsum and uranium, each of which is the subject of regulatory concern for different reasons. On the assumption that the desired sustainable outcome is food and energy security, there is a predictable point of equilibrium in respect of risk and benefit. While this point is quantifiable through the score card, it is likely to be different for each community or society. In practice, the precise point at which equilibrium between P production and consumption is both economically and socially the optimum outcome is a function of relative social attitude and absolute data.

In a society where food security is at risk, equilibrium may be evaluated differently from a society where phosphate as a potential pollutant is a primary concern. But in both instances, by applying the principle of equilibrium, the time driven weakness of the market solution, which leads inevitably to commoditization, is eliminated and a relatively sustainable outcome is achieved. The price of success in such a scenario is that the P industry will need to be significantly recapitalized, in part from the state. But as the legacy costs of the

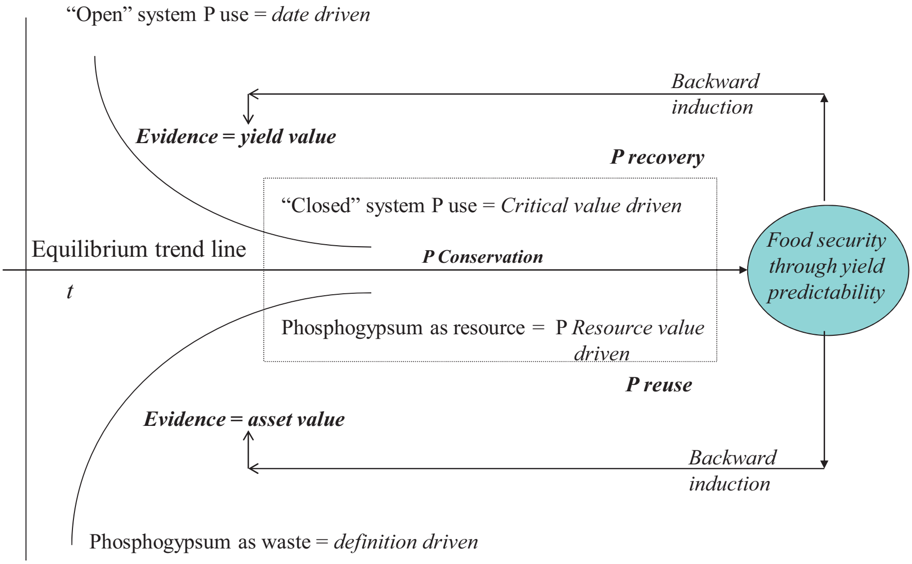


FIG.5. Backward induction: from open risk to sustainable equilibrium.

industry, as evidenced already in Florida, are very high, the choice is not between investment by the public purse or not, but rather whether the money disbursed by the public purse is deployed as an investment, on which a return can be expected in TBL terms, or whether it is simply a cost, as was the case for example in dealing with the Piney Point stack and is likely to be the case in other such situations in the future.

Using a balanced score card system may therefore facilitate the contextualization of discussions regarding risks in NORM industries against their potential benefits. The outcome is a practical and operational review of what can be done to eliminate or mitigate such risks, ensuring that actual hazard (absolute risk) is not the inevitable consequence.

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MANAGEMENT OF SCRAP METAL CONTAMINATED WITH RADIONUCLIDES OF NATURAL ORIGIN

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Abstract

During the last 10 years or so, hundreds of pieces of scrap metal contaminated with radionuclides of natural origin have been detected in Spanish scrap yards and melting facilities (about 50% of the total number of items detected). This scrap originated mainly from the decommissioning of ore processing facilities and oil and gas extraction and production facilities. The detection of these materials is a consequence of the nationwide programme established in Spain (the ‘Spanish Protocol’) to control the presence of radioactivity in scrap metal after the melting in 1998 of a high activity Cs-137 source in a melting facility. This programme has been widely implemented in Spain and the industries joining this programme (the majority of those in Spain) have installed control and monitoring systems to detect the inadvertent presence of any kind of radioactivity in the scrap before entering the plant. As part of this protocol, it is the task of the Spanish National Company for Radioactive Waste Management (ENRESA) to recover and dispose of, at suitable facilities, any material classified as radioactive waste. If the radioactivity content in the scrap is low enough, the scrap can be processed at the melting facility, with special consideration being given to instances when the radioactivity is only of natural origin. The applicable radioactivity criteria proposed by the Nuclear Safety Council have been published by the competent national authority. The values are based on recommendations of the European Commission for the recycling of metals obtained during dismantling of nuclear installations. In this paper the main characteristics of the materials detected are presented, together with the methodology for evaluating and quantifying the total activity and activity concentration. Depending on the radioactivity content, the scrap is either melted in the facility or removed as radioactive waste by ENRESA. Several operations are performed in situ to reduce the volume of radioactive waste to be removed. It is concluded that the majority of the scrap containing only radioactivity of natural origin can be melted in the facilities concerned.

1. INTRODUCTION

During the last ten years or so, several tonnes of scrap metal contaminated with radionuclides of natural origin have been detected in Spanish scrap yards and melting facilities. This scrap originated mainly from the decommissioning of ore processing facilities (other than those associated with the extraction of uranium or thorium) and facilities for oil and gas production and phosphate fertilizer production.

In oil and gas production, during the extraction and purification of crude oil and natural gas, several residues are generated (slags and deposits), as well as contaminated equipment. Some of the residues are disposed of at sea or on land, while scrap metal is mostly recycled. Contaminated equipment may also be generated by other industries.

1.1. Protocol for collaboration on the radiation monitoring of metallic materials

The detection of these materials is a part of a nationwide programme entitled *Spanish Protocol for Collaboration on the Radiation Monitoring of Metallic Materials* (the ‘Protocol’). The object of the Protocol is to establish radiological control systems for metallic materials and final products in order to detect the existence of radioactive material. This programme has been widely implemented in Spain after the accidental melting of a high activity Cs-137 source in a melting facility in 1998.

The Protocol was signed in November 1999 by all the parties concerned: the relevant Ministries, the Nuclear Safety Council (CSN), the Spanish National Company for Radioactive Waste Management (ENRESA), the trade unions and the industrial companies that recover, handle, store and recycle scrap metal. The companies that signed up to the programme have installed monitoring systems to detect the presence of any kind of radioactivity in scrap before entering the plants. They also have additional fixed and/or portable equipment to segregate and control any radioactive material detected and to control the melting products, the slag and the furnace dust.

The Protocol is complemented by a Transfer Authorisation general permit published by the Ministry of Industry and Energy in February 2000 [1], which contains the radioactivity criteria proposed by the CSN to classify the detected radioactive materials as radioactive waste. These values are based on the recommendations of the European Commission for the recycling of metals resulting from the dismantling of nuclear installations [2]. Material classified as radioactive waste is collected and disposed of by ENRESA in suitable facilities, while material with lower levels is processed at the melting facility in question. There are some technical problems still to be solved with respect to the authorization, for example the issue of low activity sources and the presence of NORM. The relevant radionuclides of natural origin (^{226}Ra and ^{232}Th) have a reference level in the general permit of 1 Bq/g, which is very low in comparison with their concentrations in many natural materials. Nevertheless, items of scrap with higher values can be melted, subject to CSN agreement, when the quantity of material involved is sufficiently low.

1.2. ENRESA activities

During this period of more than 10 years, ENRESA has carried out the following types of activity:

(a) Radiological surveillance, as required by the CSN, after other incidents occurring in melting facilities;
(b) Localization, segregation and collection of radioactive sources, also as required by the CSN;
(c) Characterization and conditioning of several types of radioactive material (sources, consumer goods, contaminated material) detected before they are removed for final disposal or returned to suppliers.

During 1998–2009, ENRESA carried out 358 interventions involving 247 instances of collection and transport at 24 melting facilities and 37 scrapyards. In all, 275 radioactive sources were collected, of which 36% were industrial sources and 64% were low activity radium sources. Additionally during this period, 2648 items containing radioactive material were detected, including sources, consumer goods (lightning rods, luminous dials) and depleted uranium (shielding and counterweights). About 41% of these items were contaminated only with NORM.

2. DETECTION OF MATERIALS

2.1. Detection process

As has been pointed out, companies subscribing to the Protocol are required to have control systems capable of detecting the presence of any type of radioactivity in scrap. In general, portal type detection equipment is used, through which the trucks transporting scrap pass. These portal monitors are normally fitted with two detection panels located on either side of the path of the truck. Each panel has one or two large plastic scintillation detectors, together with their associated electronics, the sensors detecting the passage of the vehicle, and support and protection systems. The panels also have external shielding to minimize the influence of the ambient background. The panels are located at a height suitable for screening the entire truck and at a distance sufficient to allow it to pass through unhindered (Fig. 1).

FIG. 1. Portal monitor in operation.

During periods when there is no vehicle passing through, the detectors count the ambient background. When a vehicle enters, the entry control sensor is activated and the equipment begins to measure the load. The measurement process is terminated when the exit sensor is activated, indicating that the vehicle has passed. These sensors also monitor the speed of the vehicle, which should be low. If the speed is too high for the performance of the measurement, a speed alarm is activated.

The vehicle, when passing through the measurement zone, shields the detector from background radiation, causing the counting rate to decrease (see Fig. 2). The expected background attenuation is computed by the monitoring equipment and an alarm level is set, generally at 3.5–16 standard deviations (depending on the duration of the measurement) above this modified background level. The equipment is normally set up with two or three alarm levels in order to give more information on the intensity of source.

When an alarm is generated, the procedure is to request the vehicle to pass through at least two more times. If the alarm is confirmed, the presence, position and general features of the radioactive material must be verified. To accomplish this, the vehicle is isolated and dose rate measurements are carried out in contact with the load box using portable monitoring equipment. This allows some evaluation of the radiological risk, which might be high if the vehicle is carrying an unshielded source of high activity. Once the radiological risk is confirmed to be acceptable, the vehicle is unloaded and the load inspected (see Fig. 3). The radioactive material is located and identified with the help of portable equipment,

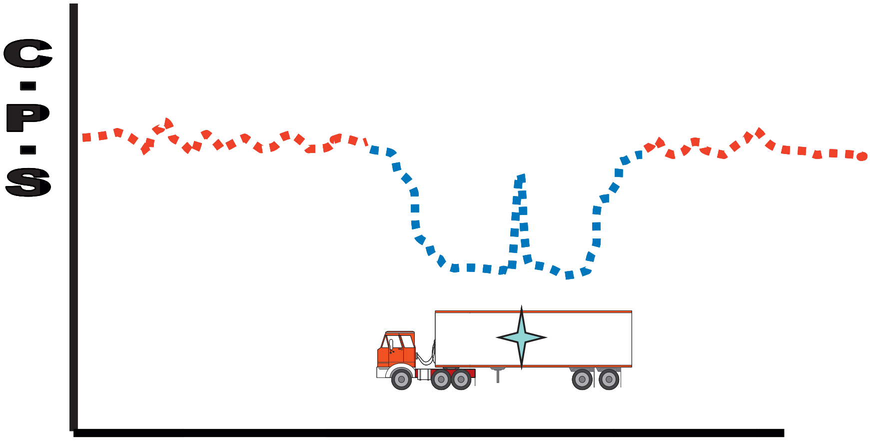


FIG. 2. Counting profile during the passage of a load.

FIG. 3. Off-loaded scrap metal.

although in certain cases the cause of the alarm is not finally confirmed. The cause may be related to the presence of large hollow items or items containing very dense material, both of which can influence the amount of shielding provided by the load and cause the calculated attenuated background to be exceeded. In certain cases, the activation of the alarm has been caused by the driver having been recently subjected to medical procedures involving radioisotopes.

Throughout the process, emphasis is placed on the need for the alarm to be confirmed in all cases, including when the level measured by the portal monitor is low, since in such instances the alarm might not necessarily have been activated by NORM posing a low level of risk. The alarm might well have been activated by the presence of a high activity shielded source, which would give rise to serious radiological consequences if it were to be melted. Similarly, there is a clear need to properly know and quantify the radiological characteristics of the material detected, in order to select and facilitate the most appropriate management approach to be followed subsequently.

2.2. Characteristics of the objects and materials detected

During the period 1998–2009 there were about 1000 cases of detection at the facilities of companies adhering to the Protocol. In many such cases, more than one type of radioactive object or material was detected. The detected items included radioactive sources with or without shielding and consumer products containing radioactive material, such as smoke detectors, lightning rods, thorium alloys and articles with luminous paint containing ^{226}Ra. There were also metallic items contaminated with NORM, with radionuclides of artificial origin such as ^{60}Co and ^{137}Cs and with depleted uranium. These items included tubing, panels, valves and compacted material.

A complete analysis of the items detected is given in Tables 1 and 2. Of the 2648 items detected, 275 were identified as radioactive sources, 64% of which were ^{226}Ra sources, generally of low activity and arising from past activities. Most of the remaining 2373 pieces (65%) were contaminated with NORM. Accordingly, the percentage of occasions on which NORM was detected was the highest (41%). In general, the radionuclide detected in the material adhering to the object was ^{226}Ra, although on certain occasions ^{232}Th was detected. A selection of NORM-contaminated objects is shown in Fig. 4. On very few occasions (1.5%) no radioactive material was found, for the reasons indicated in Section 2.1.

The dimensions of the objects varied considerably and depended on whether the scrap had previously been cut into smaller pieces. In general, the tubing varied in length from a few centimetres to 1–2 m, with a thickness of 0.2–0.5 cm, a diameter of 5–30 cm and a mass of some tens of kilograms. The panel was usually 10–30 cm long and 10–50 cm wide and had a mass also of some tens of kilograms.

From the radiological point of view, most objects exhibited a dose rate ranging from 0.3 to 1–2 μSv/h, although occasionally the dose rate reached 20–30 μSv/h. In any event, given the way in which this material was handled — with magnets and slings — the dose received by the workers was not significant. Furthermore, the total

TABLE 1. CHARACTERISTICS OF THE OBJECTS AND MATERIALS DETECTED

	Number of detections		Number of items detected	
Items contaminated with NORM:				
Objects	327	(35.9%)	1321	(49.9%)
Residues and and soil	46	(5.1%)	72	(2.7%)
Items contaminated with radionuclides of artificial origin:				
Objects	65	(7.2%)	96	(3.6%)
Residues and soil	5	(0.5%)	6	(0.2%)
Radioactive sources:				
^{226}Ra	81	(8.9%)	176	(6.6%)
Other	89	(9.8%)	99	(3.7%)
Consumer products containing radioactivity:				
Products containing Ra or Th	179	(19.7%)	625	(23.6%)
Smoke detectors	32	(3.5%)	128	(4.8%)
Lightning rods	47	(5.2%)	53	(2.0%)
Objects containing uranium	24	(2.6%)	34	(1.3%)
Containers	1	(0.1%)	1	(0.04%)
Non-radioactive items	14	(1.5%)	37	(1.4%)
Total, excluding sources	740	(81.3%)	2373	(89.6%)
Total, including sources	910	(100%)	2648	(100%)

amounts of material detected per year were not large, as a result of which the annual exposure period was very short.

The activity concentrations in the NORM-contaminating material varied over a wide range, as shown in Table 3. Occasionally, values exceeding 100 Bq/g were measured. The activity concentration of ^{226}Ra was generally higher than that of ^{232}Th. The gamma emitters of the radium and thorium decay chains were used as the basis for determining activity concentrations, with the activity concentrations of ^{226}Ra, ^{214}Pb and ^{214}Bi being measured in the case of the radium decay chain, and ^{228}Ac, ^{212}Pb and ^{208}Tl being measured in the case of the thorium decay chain. In the radium decay chain, an equilibrium factor of 0.9–1 was

TABLE 2. NORM-CONTAMINATED ITEMS

	Contribution to total number of items (%)
Objects:	
Tubing	35
Panels	4
Cylindrical filters	1
Valves	1
Drums	1
Compacted items	<<1
Safes and similar objects	<<1
Miscellaneous objects	48
Material:	
Slag	2
Refractory material	1
Residues	1
Soil	4
Other material	2

observed in 66% of the samples studied, both between ^{226}Ra and ^{214}Pb and between ^{226}Ra and ^{214}Bi. In the thorium decay chain, equilibrium was observed between ^{228}Ac and ^{212}Pb in only 27% of the cases studied, with a higher concentration of ^{212}Pb observed in 55% of cases. The equilibrium factor for ^{228}Ac and ^{208}Tl was in the range 0.3–0.4 for 55% of the samples studied.

2.3. Estimation of activity in the materials detected

The estimation of activity is normally a difficult task and is based on the measurements performed during the technical inspection of the pieces. Dose rate measurements are first used to assess the radiological risk. A spectrum is then obtained (see Fig. 5) using portable spectrometry equipment. If there is sufficient material adhering to the object, a sample is taken for subsequent analysis in the laboratory.

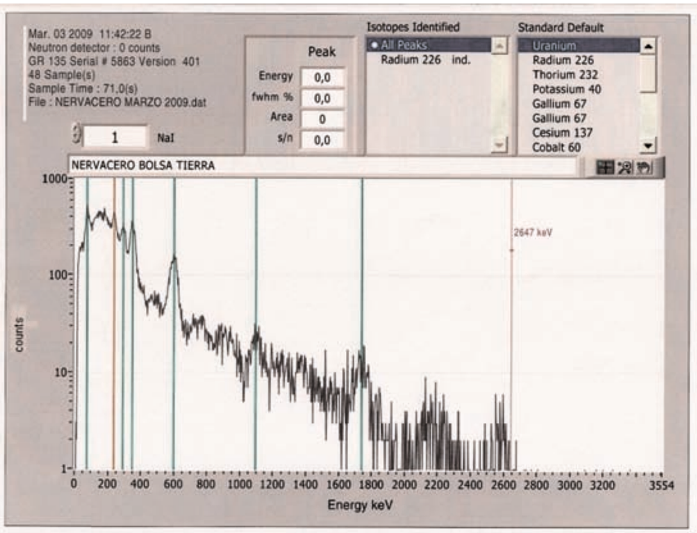
In addition to the dose rate measurements, monitoring is performed using a scintillation crystal detector (see Fig. 6) with a sensitivity much higher than that of the Geiger-Müller detector used for the dose rate measurement. This serves as

FIG. 4. Objects contaminated with NORM.

TABLE 3. DISTRIBUTION OF ACTIVITY CONCENTRATIONS IN NORM-TYPE CONTAMINANT

Activity concentration interval (Bq/g)	Relative frequency	
	^{226}Ra	^{228}Ac
<0.1	2	2
0.1–1	4	5
1–10	25	0
10–100	43	10
100–1000	45	4
1000–5000	5	1
>5000	1	0

a check to ensure that the object does not exhibit higher values that might indicate the presence of a radioactive source within it. It also serves to confirm and support any decision on when and how a particular object may be incorporated in the process when the dose rate level is close to the background value and when the spectrometry equipment gives a spectrum characteristic of the background, with no indication of an unusual energy peak.



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Neutron detector : 0 counts
GR 135 Serial # 5863 Version 401
48 Sample(s)
Sample Time : 71,0(s)
File : NERVACERO MARZO 2009.dat

1 NaI

Peak
Energy 0,0
fwhm % 0,0
Area 0
s/n 0,0

Isotopes Identified
All Peaks
Radium 226 ind.

Standard Default
Uranium
Radium 226
Thorium 232
Potassium 40
Gallium 67
Gallium 67
Cesium 137
Cobalt 60

NERVACERO BOLSA TIERRA

FIG. 5. Typical spectrum obtained for radium.

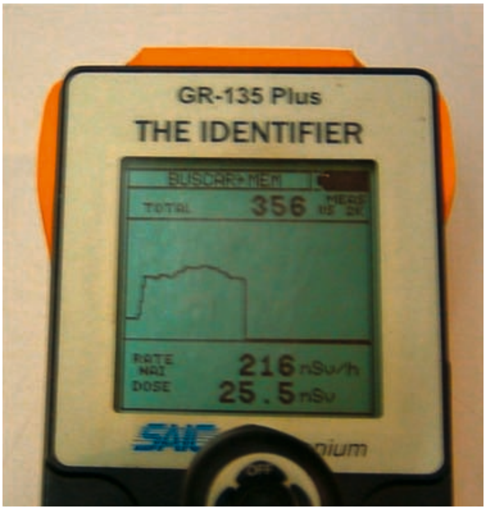

FIG. 6. Scintillation crystal detector.

The activity is estimated by calculating, using a commercial shielding calculation programme, the dose rate that would be exhibited by a radioactive source representative of the object to be assessed. The calculation is performed for a given source-to-detector distance, taking into account the isotopic characteristics, interposed materials, the geometry, and the physical and chemical characteristics. The activity is then obtained from the expression:

$$A = TD \times F$$

where A is the activity (MBq);
TD is dose rate (μSv/h);
F is the theoretical factor relating the dose rate to the activity (MBq per μSv/h).

If the measured dose rate is sufficiently high, additional dose rate measurements are made at various distances to obtain a better estimate of the activity. The factor for each distance is calculated and the activity is then calculated using the expression:

$$A = \frac{\sum_{i=1}^{n} TD_i \times F_i}{n}$$

The theoretical model to be applied will be the one closest to reality in the judgement of the technician. At times there may be several such models. Attempts are made to use the simplest model if the approach provides results that can be judged as reasonable in terms of some appropriate technical criterion.

Sometimes, the activity content of the object is estimated from the results of laboratory analysis. In this case, the estimation of the quantity of radioactive material associated with the object is based on visual inspection.

In either situation, once the activity has been calculated, it is divided by the mass of the object to obtain the activity concentration, which is then compared with the relevant reference level (1 Bq/g in the case of ^{226}Ra and ^{232}Th). If the value is higher than the reference level, ENRESA removes the material as radioactive waste. If the value is lower, a report is issued indicating that the material may be incorporated into the melting process, based on the criteria established by the CSN. Experience shows that objects exhibiting a dose rate of around 2 μSv/h or less may normally be incorporated into the process, although the measurements and calculations indicated above are carried out in all cases.

3. MATERIALS MANAGEMENT AND CONDITIONING

Objects for which the activity concentration is higher than the reference levels established in the general permit issued by the Ministry of Industry are managed as radioactive waste and sent to the ENRESA facility in Cordoba (Cabril Disposal Facility) for disposal. To enable such objects to be placed in suitable containers for disposal, it may be necessary to cut them into smaller pieces. Where possible, the object is cut open in such a way as to allow the NORM to be removed and disposed of separately as radioactive waste, thus reducing the volume to be disposed of. The decontaminated metallic parts are then incorporated into the melting process after first checking that all radioactive material has been removed.

4. CONCLUSIONS

Most of the metallic objects and material detected by the portal monitors at the entry to each metal recycling facility contain only NORM. Many of these items exhibit a very low activity concentration and may be incorporated into the melting process following a rigorous procedure to support such a decision. The application of this procedure, which includes dose rate measurements, ensures that the radiological risks to the workers at these facilities are negligible.

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TRANSPORT OF NORM

(Topical Session 3)

Chairperson

D.G. WYMER
International Atomic Energy Agency

Invited Paper

THE APPROPRIATE LEVEL OF REGULATORY CONTROL FOR THE SAFE TRANSPORT OF NORM

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Abstract

In 2006, the IAEA embarked on a Coordinated Research Programme (CRP) to examine the adequacy of the current safety standards pertaining to the transport of NORM. The CRP incorporated research studies from nine countries related to the transport of various types of NORM. The research areas provided full coverage of the subject, including exposure to both the public and workers. In addition, the CRP addressed issues of a less technical nature relating to denial of shipment and perception of harm from the transport of NORM, a subject of great interest to modern commerce. An important consideration was the appropriateness of the factor of 10 applied to the exemption level for transport of NORM not intended for the extraction of radionuclides and the factor of 30 applied to the definition of low specific activity material of the group LSA-I. The final research report, conclusions and recommendations from the CRP will be reviewed by the relevant IAEA advisory committee later in 2010. This paper provides, in the meantime, details of the research conducted, the results obtained and the preliminary conclusions.

1. BACKGROUND INFORMATION AND RATIONALE FOR THE COORDINATED RESEARCH PROGRAMME

The 1996 edition of the IAEA Transport Regulations [1] introduced radionuclide specific exemption levels in place of the former, single value of 70 Bq/g for all radionuclides combined. To avoid unnecessarily bringing ores, tailings, and backfill from large mining operations (for example, phosphate, coal, gold and monazite mining) into the scope of the Transport Regulations, the 1996 edition made provision for a factor of 10 to be applied to the normal exemption values, provided that the material concerned was not intended to be processed to extract the naturally occurring radionuclides. Even so, some mining related materials that were not previously considered radioactive material for transport purposes now became subject to specific requirements for packaging, communication, training and emergency response.

In July 2003, an international conference on the Safety of Transport of Radioactive Material took place in Vienna to address a range of important issues associated with the safe transport of radioactive material [2]. Among the topics identified for further work was the reconsideration of the applicability of the 1996 edition of the IAEA Transport Regulations [1] to NORM. The Conference suggested that the full impact of and technical basis for the factor of 10 be thoroughly researched, in order to relieve any unnecessary regulatory burden related to the transport of very low activity NORM.

As a result, the IAEA established a Coordinated Research Programme (CRP) on the Appropriate Level of Regulatory Control for the Safe Transport of Naturally Occurring Radioactive Material (NORM) to investigate this particular area of concern with regard to the Transport Regulations. A CRP schedule usually involves three Research Coordination Meetings (RCMs) about 18 months apart. Accordingly, following a preparatory meeting in November 2006, RCMs were held in April 2007, February 2008 and November 2009. The CRP report is currently being finalized. Completion of the CRP usually involves the preparation of an IAEA-TECDOC which summarizes the work performed under the CRP and includes any recommendations made by the CRP. It is envisaged that the results of the CRP could be used to revise the Transport Regulations or to incorporate new guidance into the associated advisory material addressing the transport and packaging of NORM [3]. However, recommendations from a CRP cannot be fed directly into the process for revision of the Transport Regulations. The procedures for review and revision of the Transport Regulations are started every two years. In terms of such procedures, the onus falls on one or more Member States interested in pursuing recommendations from a CRP to submit proposals for a suitable revision.

2. REGULATORY CONTEXT

The IAEA is the United Nations (UN) agency mandated “to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world” [4]. Included in this mandate is the authorization “To establish or adopt standards of safety for protection of health and minimization of danger to life and property (including such standards for labour conditions)” [5]. The IAEA first published transport regulations in 1961. Over the following decades, comprehensive reviews by Member States and international organizations were undertaken. The current edition of the IAEA Transport Regulations (TS-R-1) was published in 2009 [6]. TS-R-1 acts as a recommendation and forms the basis for other modal and national regulations, for example Recommendations on the Transport of Dangerous Goods, Model

Regulations (UN Model Regulations), International Maritime Dangerous Goods Code (IMDG Code for transport by sea) and Technical Instructions for the Safe Transport of Dangerous Goods by Air (ICAO Technical Instructions for transport by air). A companion document, Advisory Material for the IAEA Regulations for the Safe Transport of Radioactive Material (the Advisory Material), was published by the IAEA in 2008 [3].

TS-R-1 provides exemption values expressed in terms of radionuclide specific activity concentration (in becquerels per gram) and radionuclide specific total activity (in becquerels) below which the Transport Regulations do not apply. The concentration and total activity exemption levels both have to be exceeded in a consignment before the Transport Regulations apply. For the transport of NORM, exemption is defined in para. 107(e) of TS-R-1 as applying to “Natural material and ores containing naturally occurring radionuclides which are either in their natural state, or have only been processed for purposes other than for extraction of the radionuclides, and which are not intended to be processed for use of these radionuclides, provided the activity concentration of the material does not exceed 10 times the values specified in Table 2, or calculated in accordance with paras 403–407”. This is further explained in para 107.4 of the Advisory Material, which states “a factor of 10 times the exemption values for activity concentration was chosen as providing an appropriate balance between the radiological protection concerns and the practical inconvenience of regulating large quantities of material with low activity concentrations of naturally occurring radionuclides.”

The exemption levels are effectively raised by a factor of 10 for natural materials and ores “whose usefulness does not lie in the fissile, fertile or radioactive properties of those nuclides”, including materials processed by physical and/or chemical means provided the purpose was not to extract radionuclides. For the transport of NORM, the radionuclides of concern are often only those associated with natural thorium (Th_{nat}) and natural uranium (U_{nat}), provided that the parent radionuclides are in natural equilibrium with their decay products. The exemption limits listed in TS-R-1 are 1 Bq/g for Th_{nat} and for U_{nat}, therefore for NORM not intended for use of the radioactive properties the effective exemption level is 10 Bq/g.

The exemption levels given in TS-R-1 (before applying the factor of 10) were initially derived for inclusion in the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS) [7] by establishing a set of representative exposure scenarios and determining the activity concentrations and total activities that would give rise to doses to appropriate critical groups that correspond to the dose criteria for the exemption of practices without further consideration set out in Schedule I of the BSS, on the basis that the dose is of the order of 10 μSv/a or less. Para 401.4 of the Advisory Material notes that the scenarios used to derive the exemption values in the BSS were not specifically related to transport situations. However, calculations for transport scenarios were performed subsequently and it was found that the derived exemption levels were similar to the BSS values and hence, to avoid potential complications, the exemption levels derived for the BSS were adopted for the Transport Regulations.

3. THE COORDINATED RESEARCH PROGRAMME

Experts from nine countries participated in the CRP: Brazil, Canada, France, Germany, Islamic Republic of Iran, Israel, Romania, the United Kingdom and the United States of America. A wide range of materials from NORM industries were reviewed in the studies, including: tantalum ore, phosphate rock, potash, zirconium ore (zircon sands) and other materials for the ceramics industries, scales from the oil and gas industry, coal and coal ash, residues from waterworks, residues from rare earths extraction and ore and residues from uranium mines. Australia produced a report which was made available to the participants in the CRP, but did not take part. The CRP objectives and technical topics, as distributed among the participating countries, are given in Tables 1 and 2, respectively.

The participating countries conducted surveys of national industries involving transport of NORM and an assessment of doses to workers and members of the public associated with the transport of NORM. These doses were evaluated using a combination of models and measurements and were based on the work practices in place in the countries concerned. Information such as time spent driving or loading vehicles and distance from the material was used to characterize the transport operations and develop exposure scenarios. Canada, France, Germany and Israel also assessed the doses associated with the transport of NORM using a normalized modelling approach (for unit activity concentrations in the material transported) for ^{40}K, ^{238}U, ^{235}U, ^{226}Ra, ^{228}Ra, U_{nat} and Th_{nat}.

Doses were generally calculated for vehicle drivers (employees or members of the public) transporting material in a conveyance (road, rail and sea) and for individuals (employees or members of the public) loading material into a conveyance. For each of these materials, experts characterized the radionuclides, their activity concentrations and the volumes transported, as well as other aspects of the operation such as the typical loading of packages containing NORM and the types of packages used to transport NORM. Participants used the results of dose assessments to carry out an analysis of the regulatory provisions for the transport of NORM. The emphasis of this analysis varied from country to country

TABLE 1. CRP OBJECTIVES, BY COUNTRY

	Brazil	Canada	France	Germany	Islamic Republic of Iran	Israel	Romania	UK	USA
Calculate the limiting upper values for the classification of NORM as LSA-I	×								
Calculate the quantities of NORM in excepted packages, based on A_1 and A_2 values	×			×	×				
Calculate the A_2 value for NORM by the Q system	×		×						
Provide a sound theoretical basis for limits adopted by IAEA regarding LSA-I	×								
Propose new criteria for application of the Transport Regulations to NORM	×	×		×		×			
Develop a model for relating activity and dose rate	×	×		×					
Estimate doses received by transport workers and the public	×	×	×	×	×	×	×	×	×
Estimate exposure from accident scenario	×	×					×		
Examine the validity of the values for general exemption and for exempt material	×	×	×	×	×	×			×
Estimate doses received by workers during loading, unloading and storage during transit		×	×	×	×	×		×	
Consider general exemption for specific types of NORM	×	×				×			
Examine areas for radon doses				×	×	×	×		
Determine collective population doses					×		×		
Determine air dispersion factors				×			×		
Estimate background doses near sites					×		×		
Consider uniform application of para. 107(e) of the Transport Regulations to all NORM	×			×					×
Evaluate transport differences between NORM in and out of the nuclear fuel cycle	×								×

TABLE 2. CRP TECHNICAL TOPICS, BY COUNTRY

	Brazil	Canada	France	Germany	Islamic Republic of Iran	Israel	Romania	UK	USA
Characterization of NORM, such as radionuclide composition, activity concentration, shipment volume		×	×	×	×	×		×	×
Typical loading of packages containing NORM		×	×	×	×			×	×
Types of packages used to transport NORM		×	×	×	Bulk only	Bulk only	×	×	×
Dose rate measurement or calculations from packages and conveyances	×	×	×	×	×	×		×	
Characterization of transport operations, such as exposure scenarios, times, distances, intakes)		×	×	×	×	×		×	×
Measurement and modelling of worker doses		×	×	×	×	×	×	×	×
Modelling of public doses, including implications for workers not occupationally exposed	×	×	×	×			×	×	
Package performance under normal and accident conditions (extent of dispersal of contents)		Subjective only							
Consequences of transporting certain specific NORM unpackaged			×			×	×	×	×
Risk impacts due to breach of confinement of the radioactive material (modelling accident risks)	×	×							
Analysis of regulatory provisions, such as para. 107(e), LSA-I definition, unirradiated uranium definition	×	×	×	×	×	×		×	×

and included consideration of the validity of exemption levels. Some countries proposed new exemption values and considerations for a general exemption for specific types of NORM, and made suggestions for modifying certain paragraphs of the Transport Regulations (for example, para 107(e)).

3.1. Brazil

The main objectives of the Brazilian study were to establish the quantities of NORM that can be exempted from the Transport Regulations and to specify the quantities of NORM that can be transported in excepted packages. The study also aimed to provide a sound basis for the classification of NORM as LSA–I, regardless of its activity concentration above the values adopted for exemption from the Transport Regulations. To achieve these goals, a mathematical model and computer program were developed that took into account various sizes of consignment, (small to infinite size) and the U and Th decay chains in secular equilibrium, and considered both accident scenarios of the Q system combined with the trivial dose of 10 μSv (20 μSv/h for a period of 30 min) and a normal transport condition scenario with a dose limitation of 0.3 mSv/a to the driver. A summary of the results obtained showed that the most conservative scenario for the transport of NORM was considered to be the external dose to the driver under normal transport conditions, and not the accident scenarios based on the Q system hypotheses. Thus, for this most conservative scenario an average factor of 15 could be used for the exemption of NORM materials from the transport regulations, when no shielding between the radioactive load and the driver is considered. This factor is conservative and of the same order of magnitude of the factor of 10 adopted already in the Regulations. The results also pointed out that the limiting factor of 30 adopted in the Transport Regulations for the classification of NORM as LSA–I should not exist, since it is impossible when transporting NORM to reach the limiting condition of 10 mSv/h at a distance of 3 m. Finally, calculations suggested that a factor of 20 times the exemption value of natural U or Th in secular equilibrium given in Table 1 of TS-R-1 could be adopted to limit the activity concentration of NORM transported in excepted packages.

3.2. Canada

The main objectives of this study were to determine the radiological characteristics of the transport of tantalum raw materials, specifically tantalite and tin slag, and to evaluate the potential radiological exposures associated with normal transport and in the event of an accidental spill. The radiation doses received by transport workers and the public were evaluated. Chemical and physical analyses and a radiation survey was carried out on 71 shipments of material. An analysis of 67 of the shipments of tantalite and slag showed a range of about a factor of 10 in radioactivity concentrations, with an average activity concentration (^{238}U and ^{232}Th combined) of about 20 Bq/g for tantalite and about 25 Bq/g for slag. The majority (78%) of tantalite shipments and 45% of the slag shipments had concentrations exceeding 10 Bq/g.

A model showing the relationship between tantalum raw materials and the expected dose rate was developed. Based on results calculated using Microshield software, it was found to provide a consistent but somewhat conservative estimate (overestimate) of measured gamma dose rates. Exposure scenarios that considered both duration and location of exposure were established for several types of transport workers and for members of the public. Based on an evaluation of potential exposure pathways, exposure to gamma radiation was determined to be the only significant exposure pathway. Doses from exposure to spilled materials due to potential accidents were calculated and determined not to be of regulatory concern, as the resulting doses were less than 10 μSv/a.

An assessment of potential dose rates around the transport containers was conducted using the range of measured radioactivity concentrations and modelling of the associated gamma radiation doses using MicroShield software. The modelling approach overestimated the measured dose rates, primarily due to the assumption that the transport containers always carried full loads, whereas in practice the loading pattern varied. On the basis of the analyses of doses arising from the transport of tantalum raw materials described in the report there is no apparent dose related reason for an exemption value as restrictive as the current value of 10 Bq/g for these materials. No one would be expected to receive a dose above 1 mSv/a arising from the transport of tantalum raw materials. Irrespective of the exemption value selected, the dose assessments described in the report provide assurance to the tantalum industry and to its shippers that the doses arising from the transport of tantalum raw materials are low and well within international limits for both transport workers and members of the public. Using conservative assumptions and on the basis of a 0.3 mSv/a reference dose, an exemption value of at least 30 Bq/g is considered appropriate for the transport of tantalum raw materials.

3.3. France

France aimed at calculating A_2 values for the materials whose values are currently listed as ‘unlimited’, and exemption values for larger quantities of material (~20 t). The study also examined the validity of the 10 mg limiting intake and addressed the A_2 values derived for accident conditions. The study took account of different transport conditions, for example, whether the material is in drums or bags. The results included a dose assessment based on the normalized scenarios and an evaluation of A_1 and A_2 values for NORM according to the Q system. In addition, one industrial activity (coal combustion in power plants handling NORM) was assessed. A study of the workplaces of the drivers was performed. Dose assessments were based on realistic scenarios, and the results of calculations are in good agreement with measurements. For a normalized ^{226}Ra concentration of 1 Bq/g, the results showed that the annual doses received by a vehicle driver and a forklift driver were 330 and 500 µSv, respectively. Deposits in pipes were also investigated. The ^{226}Ra concentration in these deposits can reach high values, especially for pipes in the phosphate industry, where activity concentrations of up to 1600 Bq/g are reported. The dose received by a scrap metal worker specialized in dismantling industrial facilities was calculated and found to be in good agreement with the measurements. The doses for a normalized ^{226}Ra concentration in the deposit of 1 Bq/g were 0.13 µSv/h for the driver, leading to 0.39 µSv for the driver for one transport of 3 h and 100 µSv/a for a full-time activity of 800 h/a. Industrial workplace studies from different types of industrial facilities such as coal combustion in thermal power plants, treatment of tin, aluminium, copper, titanium, niobium, bismuth and thorium ores, and production of refractory ceramics were done. In those evaluations, the doses received by operators were assessed. Altogether, 3800 measurements were performed on 475 samples of material. The transport of uranium ore from mines to concentrating plants was studied. The material being transported was viewed in terms of activity concentration, density and dust inhalation. Workers such as the truck driver, fork-lift truck driver, truck loader and worker on a stack of material were considered. The study concluded that density has a low impact on the dose rates and the external dose rates received by each of the four workers are of the same order of magnitude. Realistic exposure scenarios for the truck driver and the fork-lift truck driver were evaluated and showed that transporting uranium ore with an activity concentration of 1 Bq/g would result in an annual dose of about 100–150 µSv, while transporting radium-containing waste with an activity concentration of 10 Bq/g would result in an annual dose of about 1–1.5 mSv.

For NORM not intended to be processed for the use of the radionuclides contained within it, a factor of about 10 could be assumed to take into account the variability of the activity concentrations in the loads transported throughout the year. Taking into account that factor of 10, the transport of an ore containing U_{nat} at 10 Bq/g, not intended to be processed for the use of the radionuclides, the annual dose would be about 100 µSv.

Some materials, whether in equilibrium or not, can lead to an annual dose higher than 1 mSv. For example, an employee dealing with the loading and transport of baddeleyite (raw material with a uranium activity concentration of 7 Bq/g) can receive an annual dose greater than 1 mSv in about 725 h only by external exposure. Moreover, an employee handling a bulk bag containing sand used for underground water filtration (^{226}Ra: 3.7 Bq/g; ^{228}Ra: 3.3 Bq/g) can receive an annual dose of 1 mSv in less than 1000 h only by external exposure. It is emphasised that this activity concentration in the case of radium (7 Bq/g) is much lower than the 100 Bq/g exemption level for a material not intended to be processed for the use of its radionuclides. Compared with the exemption level of 10 µSv/a, the values of the assessed doses seem to be high.

3.4. Germany

The overall objective of the German contribution was to review and categorize the most important materials containing radionuclides of natural origin; to review, analyse and evaluate the radiation exposure resulting from the shipment of NORM and the expected exposure of the shipment staff and the population; to develop evaluation criteria and safety requirements to provide adequate safety standards for the transport of NORM; and to develop procedures for determining the criteria for exempt material and exempt consignments for transport according to the Transport Regulations for all types of NORM.

Finally, on the basis of the results of the dose calculation for the transport of NORM, the following recommendations are given as far as the proposed dose criterion of 0.3 mSv/a for transport personnel is accepted:

(a) For bulk transport of NORM in equilibrium, a five-fold activity concentration factor for exempt material meets this requirement irrespective of the type and use of such material.
(b) Accordingly, para 106 (e) could be amended as follows:
  (1) Delete all references to the intended use ("… other than for the extraction of the radionuclides, and that are not intended to be processed for use of these radionuclides,…").
  (2) The last part of the sentence in para 106 (e) with the reference to paras 401 (b) to 406 should be replaced by a new paragraph which contains the exemption levels for natural radionuclides only, namely:
    - In the case of equilibrium, the activity concentration for exempt material is 5 Bq/g for U_{nat} and Th_{nat};
    - In the case of non-equilibrium, the activity concentration for exempt material is to be calculated using the formula in para 404 of TS-R-1, with the following upper bounds:
      — 15 Bq/g for ^{226}Ra and 10 Bq/g for ^{228}Ra;
      — 100 Bq/g (reflecting a factor of 10) for ^{210}Pb and ^{210}Po not in equilibrium, regardless of the 50 Bq/g upper bound obtained by applying the formula in para 404 of TS-R-1.

3.5. Islamic Republic of Iran

The study examined bulk shipments of phosphate rock from Morocco and Jordan. Assessments were carried out on the radiological impact of NORM in the phosphate, zircon and uranium industries. Dose rates from material and exposures to a variety of workers involved in vessel discharge, loading and unloading, cleaning and transport operations were measured. Uranium ore bulk shipments were assessed from the mine site through the processing plant to the transport routes, including various dose assessments. Various ores are blended to achieve 10 Bq/g of ^{238}U with low levels of ^{232}Th and ^{40}K. The truck driver and loader operator were estimated to have received effective doses of 0.062 and 2.07 mSv/a, respectively. It was noted that the dose from external exposure inside the cabin was four times higher for the loader as opposed to the truck driver, owing to the loader being closer to the phosphate rock stockpile and for a longer period of time. Data were collected on types and volumes of shipment, the radionuclide content of the materials, the types of packaging and vehicles used, the dose and dose rates around the packages used to transport these materials and the details of the transport operations for each type of material. Data were presented for bulk transport of phosphate rock. For each shipment, the range of radionuclide concentrations in various samples of phosphate rock was measured. The occupational exposure scenarios for an exposed person in the bulk transport of phosphate rock are mentioned and in each scenario the annual exposure time was estimated. Measurements of external gamma, short lived alpha emitting particles and long lived radionuclide exposure pathways were taken. The results of the dose assessments for occupational scenarios for bulk unpackaged phosphate rock were presented. In this study marine transport and loading of ships were not included; only off-loading of the ships and storage at the end of the transport route were considered. The results of the dose assessments for occupational scenarios were presented. The dose during off-loading (discharge) in all bulk transport scenarios was less than 20 μSv per shipment. The activity concentrations in uranium ore from the Islamic Republic of Iran were measured and the dose received by workers from transport of uranium ore was estimated and presented in the final report.

3.6. Israel

The study estimated the occupational exposure during activities related to the transport of potash, phosphate rock and phosphate fertilizer. These materials are transported on a bulk scale (a few million tonnes per year) in an unpackaged form. Measurements were conducted at the loading stage of the phosphate and potash products and included the radionuclide content of the phosphate and potash products, the airborne radionuclide concentrations at the loading facilities and the airborne dust size distribution. Based on these measurements, the total dose rate for a loading worker was estimated. Based on an assumption of the total annual working hours the annual dose received by a loading worker was estimated and compared with the relevant dose limits. The annual dose received by loading workers at the phosphate and potash facilities was estimated to be less than 0.3 mSv. It was assumed that the dose received by members of the public as a consequence of this work was less than 10 μSv per year. Para. 5.12 of IAEA Safety Guide RS-G-1.7 [8] states that for activity concentrations that exceed the radiological criteria for regulation by several times (for instance, up to 10 times), the regulatory body may decide, in line with the graded approach to regulation, that the optimum regulatory option is not to apply regulatory requirements and that the mechanism for giving effect to such a decision could be to grant an exemption. It was found that the concentrations of phosphate and potash products were less than 10 times these radiological criteria, implying that they would be candidates for exemption. The dose assessment for phosphate and potash loading workers indicates that the values given in the Transport Regulations for exempt quantities in transport, when subject to the factor of 10, are well suited for loading activities of phosphate and potash.

3.7. Romania

This study examined the disposal and transport of tailings from the Crucea uranium mine. The potential risks and radiological consequences associated with the transport and disposal of the material were identified and evaluated. Tailings sites were investigated in order to estimate doses received by members of the public from inhalation of radon, both under present conditions and in the future. This involved air concentration modelling, for example, modelling of long range transport, which required sophisticated models, comprehensive meteorological data and extensive set-up effort. Estimates were made of the radon source term, population densities, doses received by the exposed population, the background dose and the normalized tailings surface area. It was estimated that the effective dose received by workers did not exceed 20 mSv/a. The dose rate from external radiation was 0.5–5 μSv/h. The maximum effective dose received from radon was estimated to be 5.88 mSv in the surrounding area and 15.50 mSv at the tailings site. The average dose was estimated to be less than 0.2 mSv/a. The annual effective dose from all environmental pathways was estimated not to exceed 1.4 μSv/a.3.8. United Kingdom

This study reviewed the transport of materials containing naturally occurring radionuclides in the United Kingdom and where appropriate the radiological impact of these transport operations was assessed. Firstly, data on activity concentrations of naturally occurring radionuclides in material typically transported in the UK were collected and, secondly, the radiation exposures that may result from the transport of NORM in the UK were estimated. Coal, coal ash, iron and steel production, building materials, potash, phosphate rock and fertilizers, ores and mineral sands, and wastes from the oil and gas industry and china clay industry were all surveyed.

3.8. USA

The research undertaken in the USA included evaluation of the inconsistencies in the application of the exempt activity concentrations, particularly as they are applied on the basis of the intended use of the material being transported (for example, paragraph 107(e) of the Transport Regulations). It was concluded that the ‘10 times’ provision of paragraph 107(e) is consistent with the IAEA’s common practice of relaxing radionuclide exemption concentrations within cautious bounds to achieve a balance between practical issues and radiological concerns. Analyses based on realistic transport scenarios indicated that, in cases where the 10 times provision is applicable, the maximum annual dose from unregulated transport of natural uranium or thorium would generally be substantially less than the IAEA’s ‘practical dose constraint’ of 1 mSv. Realistic transport scenarios were identified in which the provisions of paragraph 107(e), together with the rounding methods used to establish the exemption values, led to exemption values differing by two orders of magnitude for two materials that emit the same types and energies of radiation and deliver the same dose per unit activity concentration to the person presumed to receive the highest dose. This is inconsistent with the principle that the exemption values should be risk-based.

With respect to the special provisions in paragraph 107(e), regarding the ‘prior or intended use’ (PIU) restriction, it was concluded that such a provision in paragraph 107(e) is not justified and should be removed. If exemption values are to be risk informed, they should be based on dose implications, not on the prior or intended uses of the material being transported. Consequently, allowance of a ten-fold increase in the exemption values for natural material and ores containing naturally occurring radionuclides should be applied to all such material, regardless of their past or intended use. If paragraph 107(e) is modified to eliminate the ‘intended use’ clause, it will also be necessary to remove a corresponding clause from the definition of LSA-I material. This definition includes “uranium and thorium ores and concentrates of such ores, and other ores containing naturally occurring radionuclides which are intended to be processed for the use of these radionuclides”.

The research also involved the measurement and estimation of doses associated with the transport of uranium ore and other NORM and the treatment of progeny in Footnotes (a) and (b) of Table 2 in the Transport Regulations. Based on evaluations, it was recommended that the footnotes should be revised to read as follows:

> “If a radionuclide is listed with a footnote (b) for its exemption values then it need not also be listed with a footnote (a); consequently, radionuclides with a footnote (b) should have the footnote for their A values changed from (a) to (b). This would indicate the same physical information was used in deriving the limits. Future efforts to rationalize the treatment of daughter products in the two calculation systems (A values and exemption values) should be based on ICRP recommendations.”

4. CONCLUSIONS

At the 3rd RCM in November 2009, the following conclusions were agreed upon:

(a) The doses calculated for workers involved in transport operations (drivers and loaders) were found to be within the range described in the regulatory context.

(b) The doses received by the general public were at least an order of magnitude lower.

(c) The basic exemption level of 1 Bq/g for U_{nat} and Th_{nat} was appropriate and the ‘10 times’ provision for NORM, as defined in para. 107(e) of the 2009 edition of the Transport Regulations [6], was both appropriate and necessary. The 10 Bq/g value for ^{40}K may be too restrictive given the natural ratio to stable potassium.

(d) The ‘10 times’ provision for NORM given in para. 107(e) of the Transport Regulations should be made clearer to ensure its proper application. Options include the addition of a footnote to the entries for ^{40}K, Th_{nat} and U_{nat} which would refer to para. 107(e).

(e) The need to apply para. 405 of the 2009 edition of the Transport Regulations for materials which are not in equilibrium should also be made clearer. This is best illustrated by radium which has been separated from its parent chain, where the 10 Bq/g exemption value for ^{226}Ra and ^{228}Ra may be too high when not applying the rule for mixtures.

The CRP report is being finalized and will be reviewed by the relevant IAEA advisory committee in June 2010.

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TRANSPORT OF NORM IN THE PORT OF ANTWERP: FROM MEGAPORTS TO A SPECIAL PURPOSE MEASUREMENT METHODOLOGY

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Abstract

When nuclear inspections are conducted in the port of Antwerp, many alarms are triggered by NORM. This paper presents an overview and preliminary results of a study taking place in the port of Antwerp and at NORM handling companies. The study aims to get an overview of NORM transport in the port of Antwerp, including quantity, destination, dose rates and activity concentrations. Furthermore, a methodology is being developed to obtain an in situ estimation of the activity concentration of material in containment systems typically used in industry (shipping containers and bulk bags). The preliminary results of the assessment of this methodology are discussed.

1. INTRODUCTION

Ninety per cent of the world’s trade happens via container cargo shipped in and out of seaports. If left unprotected, the seaports could be subject to nuclear smuggling to facilitate terrorist attacks with nuclear technology such as nuclear weapons and ‘dirty bombs’, which combine radioactive material with conventional explosives. That is why it is important for security around the world that cargo shipped through seaports is screened for hazardous materials to prevent smuggling in container traffic.

The Megaports Initiative [1, 2] started in 2003 and teams up with other countries to enhance their ability to screen cargo at major international seaports. The government of the United States of America sponsors the worldwide installation of nuclear detection equipment in ports. The Belgian government agreed to such an installation in the port of Antwerp in November 2004 [1].

Nuclear inspections consist of several phases. In the primary phase, all containers are inspected by radiation portals. If the radioactivity of the load is above a certain threshold, the portals will create an alarm. However, this will not give further information about the nature of the load. As a result, the container will be blocked and the manifest information will be collected. If the container contains NORM, it will be released if the radiation profile corresponds to a homogeneous load and when the threshold for that particular material has not been exceeded. Otherwise, a secondary inspection is required.

The European Commission is currently recasting five Council Directives with respect to natural radiation sources.[1] The goal is to define a uniform approach towards NORM for adoption by individual Member States. One of the new elements is the construction of a 'positive list' of industrial activities in the non-nuclear sector that may be subject to notification. As described in publications by UNSCEAR [3] and the International Commission on Radiological Protection (ICRP) [4], an acceptable dose rate threshold can be related to the activity concentration of material. As a result, the activity concentration can be used to determine whether regulatory authorities have to be notified with respect to NORM.

These recent developments were the basis of a new project, 'NuTeC–NORM', supported by the European Regional Development Fund (EFRO) and the Ministry of the Flemish community. It was initiated to gain insight into this matter of NORM in non-nuclear industrial activities in Belgium. One of the major goals of this project is the identification of NORM in a shipping container, followed by an estimation of the activity and activity concentration of each naturally occurring radionuclide identified. On the basis of this information, the construction of a database of NORM traffic passing through the port of Antwerp is planned. The practical implementation of the new directives requires a technique to determine the activity concentration of NORM. Since the activity concentration has to be measured in all the products, by-products and residues in an industrial process, a quick and easy methodology is mandatory. Nowadays, the

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[1] The Basic Safety Standards Directive (96/29/Euratom), the Medical Directive (97/43/Euratom), the Directive on High Activity Sealed Sources (2003/122/Euratom), the Directive on Outside Workers (90/641/Euratom), and the Directive on Public Information (89/618/Euratom).

activity concentration is usually measured using sample analysis which is a cumbersome, time and money consuming task. Another major goal of the NuTeC–NORM project is to develop a quick and easy to use methodology to estimate the activity concentrations in industrial settings.

2. MEASUREMENT EQUIPMENT AND METHODS

2.1. Equipment located in the port of Antwerp

2.1.1. Equipment for primary inspection

The primary inspection consists of a container passing through a radiation portal monitor, generating an alarm expressed in terms of 'sigma' for gamma radiation (where 1 sigma equals the square root of the background radiation level) and counts per second for neutron radiation. The portals are placed at the gates of the discharging quays. The portals contain four plastic scintillation detectors for gamma radiation and four ^{3}He detector systems for neutron radiation. The interpretation of the results can lead to three legally recognized possibilities: the transport is licensed, the measurements are erroneous (for instance, due to a sudden fluctuation in background or a defective detector) or the content of the shipment is NORM below an acceptable limit for the specific substances present. If none of these possibilities is true when an alarm occurs, a secondary inspection is mandatory.

2.1.2. Equipment for secondary inspection and tertiary inspection [1, 2]

When the primary inspection at the quay requires a secondary inspection, the load is sent to a 'central alarm station'. The first part of the secondary inspection consists of pulling the load through a tunnel containing a set of inspection equipment as shown in Fig. 1.

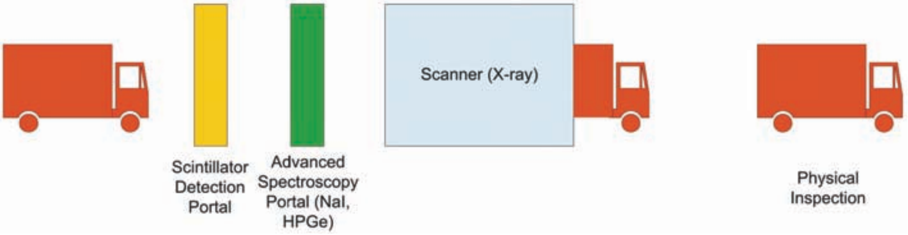

FIG.1. Consecutive steps for secondary inspection.

First the load passes a more sensitive scintillation detector and an ‘advanced spectroscopy portal’ equipped with NaI and ^{3}He detectors. The load is then pulled through a scan tunnel in order to construct an X ray image of the content of the container. In this way the content can be compared with the expected content according to the manifest.

Afterwards, a physical inspection may be performed. This inspection is conducted by a radiation expert. Various measurement devices are used to inspect the load: a gamma counter, a dose rate monitor and multi-channel analysers (NaI(Tl) and HPGe). With these measurements, the dose rates and the radionuclides present in specific parts of the shipment can be determined.

In the tertiary phase, local government officials are warned when there is no logical and legally acceptable explanation for the level of radioactivity. The inspection will be performed by radiation experts, mostly consisting of reviewing the available data. In Belgium the authorized agency is the Federal Agency for Nuclear Control (FANC) with, in most cases, NuTeC workers performing the radiation expert function up until May 2008. Soon it became clear that the greater part of the alarms were generated by containers filled with NORM.

2.2. Development of method and equipment in the NuTeC–NORM project

A new methodology provides a technique and set of tools to determine the activity concentration of NORM radionuclides using a relatively cheap NaI (Tl) 2 × 2 inch (or sometimes the more expensive LaBr 2 × 2 inch) probe based multi-channel analyser (MCA) connected to a portable tablet PC. (For comparison purposes a germanium based probe can be used, but such detectors are usually much more expensive, less portable and have a limited battery life span because of the need for electrically powered cooling.) The multi-channel analyser (scintillation detector) is positioned in contact with the container and the device is controlled by the software on the portable tablet computer. A spectrum is automatically recorded and analysed. Finally, an estimation of the activity concentration is provided to the user.

The software on the tablet PC was designed taking ease of use into account. Because customs officers and, in a later phase, people working in industrial facilities have to work with the tool, the measurement process has to be fully automated and feedback given in a comprehensive manner. The software automatically records the spectrum and performs the analysis. The result is presented to the user in a way that the user can decide on the appropriate course of action.

3. RESULTS AND DISCUSSION

In this section, the preliminary results to date of the NuTeC–NORM project are discussed.

3.1. Screening of containers: Establishment of a database

Since the Megaports project proved that many shipments in the port of Antwerp were carrying NORM, it was decided that this location would be an excellent starting point to perform a study of NORM in Belgium. First, a database was established to capture information gathered in secondary inspections with respect to NORM. In this way, it was possible to identify several companies importing goods involving the presence of NORM. Furthermore, the database includes results of the physical inspection, such as dose rate measurements, recorded spectra and sample analysis results. At the moment, the database of the secondary inspections is being linked to the primary inspections. In this way, customs officers will be able to consult historical information to compare inspections with previous cases.

As a first step, zircon has been studied. Zircon is a sand consisting mainly of zirconium silicate ($ZrSiO_4$). The largest end use of zircon is as an opacifier in the manufacture of ceramic based products, including tiles, sanitary ware and table ware. A rapidly growing sector for the use of zircon is the production of zirconia (zirconium oxide), zirconium based chemicals and zirconium metal. Other main end use markets for zircon include refractories, foundries and the manufacture of television screens [5, 6]. Zircon sand is produced mainly in Australia, South Africa and China, as shown in Table 1.

All mineral sand contains radionuclides of natural origin, primarily those in the uranium and thorium decay series. The concentrations of these radionuclides are low, but significantly higher than those in normal rocks and soil [8, 9]. A wide range of ^{238}U and ^{232}Th activity concentrations in zirconium minerals is reported in the literature, but the activity concentrations in commercially exploited zirconium minerals are less variable and tend to be at the lower end of the range. Most zircon currently produced has activity concentrations of 1–4 Bq/g for ^{238}U and 0.5–1 Bq/g for ^{232}Th; although higher activity concentrations in commercial zircon have been reported, especially from some lesser producing countries where in many cases the zircon is contaminated with monazite (see Table 2), such material tends to be avoided by most current zircon producers [9].

During the period May 2007 to December 2009, sigma values for a large number of containers passing through the port of Antwerp were measured and recorded in the database. The overall average sigma value was 29, with a standard deviation of 42.53. The sigma values associated with zirconium minerals in

TABLE 1. WORLDWIDE PRODUCTION OF ZIRCON (From Ref. [7])

	Production (t)	
	2007	2008
Russian Federation[a]	7135	7000
Ukraine	35 000	35 000
Mozambique	26 347	6552
South Africa	388 800	404 000
United States of America	121 000	122 000
Brazil	26 739	27 000
China	140 000	140 000
India	24 000	24 000
Indonesia	111 000	65 000
Malaysia	7393	984
Sri Lanka	381	1447
Thailand	1023	—
Vietnam[b]	22 000	24 000
Australia	600 000	550 000
World total	1 511 000	1 407 000

[a] Including caldasite rock containing zircon and baddeleyite.

[b] Conservative estimates, based on exports.

TABLE 2. ACTIVITY CONCENTRATIONS IN COMMERCIAL ZIRCON, BY COUNTRY OF ORIGIN (from Ref. [9])

	Activity concentration (Bq/g)		
	^{238}U	^{232}Th	^{40}K
Australia	1–5.8	0.3–1.9	0.05–0.7
China	14.4–14.7	8.0–8.2	2.2–2.3
India	1.6–6.3	0.38–0.67	
Malaysia	13–50	2.2–88	
South Africa	2.8–7.8	0.5–1.1	
United States of America	1.9–4.0	0.1–0.6	

containers over the same 2½ year period were significantly higher (see Fig. 2), with an average of 279 and a standard deviation of 160.39. Out of a total of 493 measurements, 337 were in the range 150–300. The sigma value also appeared to depend to some extent on the type of zirconium mineral in the container, as shown in Table 3. For instance, the sigma values associated with consignments designated as ‘zirconium silicate’ were typically about twice those associated with consignments designated as ‘zirconium dioxide and baddeleyite’ and ‘zirconium sands’.

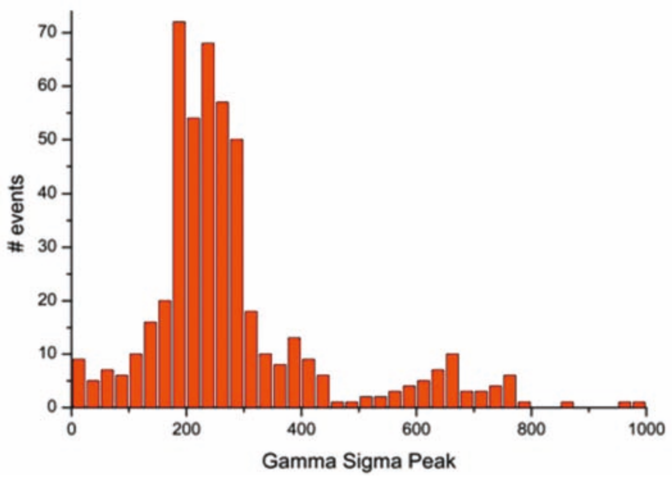

FIG. 2. Distribution of gamma radiation measurements associated with zirconium minerals in containers in the port of Antwerp (May 2007–December 2009).

TABLE 3. SIGMA VALUES FOR VARIOUS TYPES OF ZIRCONIUM MINERAL

	Number of measurements	Average sigma value	Standard deviation
Zirconium dioxide and baddeleyite	77	207	49.55
Zirconium silicate, $ZrSiO_4$	45	539	185.86
Zirconium sands	45	251	38.85
All zirconium minerals	493	279	160.39

3.2. Application of the new detection methodology in an industrial setting

A case study was conducted at a company handling zircon sand from Australia. This company has a range of container terminals at seaports and inland and stores and handles sugar, fertilizer, chemicals, minerals, iron and steel and wood products. The handling comprises bagging, repacking, sieving, sifting, weighing, mixing and conditioned storage in contamination free warehouses. The equipment and methodology used for gamma ray measurement is described in Section 2.2. For transport situations, it is very useful to measure directly on the shipping containers. In an industrial environment, however, there are different kinds of packaging with very different dimensions. The measurements were carried out on bulk bags, each having a mass of 2 t when full, with the detector positioned on the top surface for a period of 900 s. To verify the repeatability, ten such measurements were made without moving the position of the detector. The activity concentrations of ^{238}U and ^{232}Th were determined indirectly from activity conentrations of their progeny, assuming decay chain equilibrium. The accuracy of the method was checked by comparing the results with the results from an accepted method based on sample analysis.

The results of a set of 10 measurements on a 2 t bag of zircon originating from Australia are shown in Table 4, indicating that the method provides good reproducibility and accuracy for this particular sample.

4. CONCLUSIONS AND OUTLOOK

In future work, the challenge will be to improve the method for determining the activity concentration of ^{238}U. Application of the method will be extended to other types of NORM and customized for different measuring geometries in an industrial setting.

TABLE 4. MEASUREMENT RESULTS ON A 2 t BAG OF ZIRCON

	Activity concentration (Bq/g)		Relative error (%)
	Mean	Standard deviation	
^{238}U	4.07	0.84	+109
^{232}Th	0.51	0.05	+3
^{226}Ra	2.09	0.07	–2

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TRANSPORT OF NORM IN FRANCE AND THE DOSES INVOLVED

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Abstract

The transport of materials, products or waste containing radionuclides of natural origin can lead to significant exposure of workers. Exposure due to the handling and storage of NORM is taken into account by French authorities through a specific regulation, the Ministerial Order of May 25, 2005. This regulation requires operators to assess the effective doses received by workers. Based on data provided by operators in nearly 90 studies, IRSN compiled a review of the effective doses in excess of the natural background due to the transport of NORM. IRSN also constructed standard exposure scenarios defining the characteristics of the transport (the quantities transported and the shielding) and the characteristics of the exposure (such as the ambient dust concentration, the distance from the material, the transport duration and the number of transport operations per year) and then compared this with the collected data. It is concluded that waste generally contains the highest activity concentrations, that among all the radionuclides ^{226}Ra needs specific control and that the transport of NORM can lead to significant effective doses.

1. INTRODUCTION

The transport of radioactive material is subject to the IAEA Transport Regulations [1]. However, some substances with low activity concentration, the transport of which results in very low doses being received by workers and the public, are exempted from the Transport Regulations. In addition, natural materials and ores containing radionuclides of natural origin which are either in their natural state, or have only been processed for purposes other than for extraction of the radionuclides, and which are not intended to be processed for use of these radionuclides, may be outside the scope of application of the Transport Regulations. The determining activity concentration in such cases is 10 times the exemption level (see para. 107 (e) of Ref. [1]). Examples include material that contains uranium but is not used in the nuclear fuel cycle.

In 2005, the IAEA launched a coordinated research programme on the appropriate level of regulation for the transport of NORM. ASN and IRSN cooperated in this study, which was carried out over a period of four years. To examine the adequacy of the Transport Regulations for the transport of NORM, IRSN and ASN conducted a study on the main types of NORM transported in France, comprising:

(a) An overview of the types of NORM transported;
(b) A dosimetric study of workplaces linked to the transport of NORM.

2. NORM TRANSPORTED IN FRANCE

NORM transported in France can be divided into two main categories:

(1) Uranium used in the front end of the nuclear fuel cycle, that is, before enrichment. Transport involves mainly uranium concentrate (yellow cake) and the by-products of uranium extraction (purified uranium and waste).
(2) The materials used in conventional (non-nuclear) industries. These industries are very diverse (see Section 2.2.1). They use raw materials containing low concentrations of radionuclides of natural origin. IRSN reviewed 88 studies conducted in French industries with elevated levels of natural radioactivity, involving 475 samples of raw materials and approximately 3800 measurements.

2.1. NORM used for its radioactive properties

2.1.1. Uranium

Uranium mines are no longer in operation in France. The ores previously mined were low grade, with a uranium content of <1 kg/t and a typical ^{238}U activity concentration of about 13 Bq/g. The ore was concentrated into yellow cake in uranium extraction plants located close to the mines. Since the closure of the last of the French mines in 2001, uranium has been imported into France in the form of yellow cake containing about 75 wt% uranium. The uranium extraction process severs the uranium decay chain such that freshly produced yellow cake contains only the radionuclides ^{234}U, ^{235}U and ^{238}U. However, by the time the yellow cake is transported, the immediate progeny with very short half-lives will have grown back into equilibrium with their parents. The combined activity concentration of ^{234}U, ^{235}U and ^{238}U in yellow cake (without considering the activity concentrations of their short lived progeny) is about 20 000 Bq/g. Since the activity concentration of yellow cake is much higher than the criteria studied in this report, the regulatory provisions applicable to the transport of yellow cake have not been examined.

2.1.2. Thorium

Some products contain thorium extracted as a by-product of the rare earths extraction process. The activity content of such products is not significant.

2.2. NORM not used for its radioactive properties

2.2.1. Overview of the materials likely to be transported

NORM, which is not used for its radioactive properties and which is transported in France, comes mainly from the following industries:

(a) Coal combustion in thermal power plants;
(b) Processing of ores for the extraction of tin, aluminium, copper, titanium, niobium, bismuth and thorium;
(c) Production of refractories, glass making, foundries and steel production;
(d) Poduction and use of thorium compounds;
(e) Production of zirconia and baddeleyite, and casting and metallurgical activities;
(f) Production of phosphate fertilizers and phosphoric acid;
(g) Extraction of rare earths and the production of pigments;
(h) Treatment of underground water for general human consumption or bottled mineral water;
(i) Health spas;
(j) Oil and gas extraction.

2.2.2. Densities and activity concentrations of the material transported

Information on densities and activity concentrations is given in Table 1. The information on densities was obtained from the industries concerned, from material safety data sheets provided during the course of the studies and from the internet site www.mineralinfo.org. With regard to activity concentrations, IRSN

TABLE 1. NORM IN FRENCH NON-NUCLEAR INDUSTRIES

Industry	Density	Activity concentration (Bq/g)		
		Range	Median	Average
Aluminium	2.6–4.1	0.0052–7.316	0.165	0.301
Coal combustion	0.8–2.5	0.00045–18.03	0.11	0.324
Drinking water treatment	1–2	0.00003–7.63	0.17	1.433
Kaolin	0.4–2.3	0.00004–32.12	0.115	3.617
Titanium dioxide	1.4–4.8	0.0051–543.4	0.24	37.109
Rare earths	2.3–2.9	0.14–4.73	0.57	1.366
Thorium	18.8[a]	0.1–3022	9	24.088
Zircon	1.5–4.5	0.026–318.6	0.9	7.82
Fertilizer	0.9–2	0.002–15.12	0.463	1.551
Glass	0.04[b]–6.5	0.0033–19.74	0.07	0.445
Refractory products	1.1–5.3	0.00001–70.3	0.23	0.732

[a] Single value.

[b] The low densities are found in glass fibre products.

distinguished between raw materials, final products and waste (solid waste, sludge and dust). The activity concentrations were found to vary over wide ranges. More detailed information can be found in Ref. [2].

2.2.3. Dust concentrations

The measured dust concentrations vary considerably across different industrial sectors and sometimes at the same workplace on the same day. In workplaces related to transport activities, the dust concentrations vary from negligible values up to 3.5 mg/m^3. Higher dust concentrations (up to 13.5 mg/m^3) can be found in workplaces not related to transport.

3. COMMITTED DOSES FOR THE TRANSPORT OF NORM

3.1. Calculation of dose rate per unit activity concentration

For each radionuclide or group of radionuclides, IRSN calculated the dose rate (that is, the dose received in 1 h) to which a worker is exposed when involved in the transport of a cargo with an activity concentration of 1 Bq/g, where this value applied to (a) uranium isotopes combined, (b) thorium isotopes combined, (c) ^{40}K, or (d) each group of radionuclides. In the case of uranium, activity concentrations of 0.98 Bq/g for ^{238}U and 0.0227 Bq/g for ^{235}U were chosen. In the case of thorium, an activity concentration of 1 Bq/g for ^{232}Th was chosen. This approach, which considers only the activity of the parent radionuclide, corresponds to that followed in the Transport Regulations.[1]

3.1.1. External exposure

The workplaces studied were those relevant to the following types of worker: a truck driver, an employee supervising the loading of a truck from a silo, a forklift driver handling a bulk bag and a worker walking on a pile of bulk material. The dose rate calculations were performed using the computer code MicroShield version 5.0 [3].

Truck driver

The scenario considered is the following:

(a) The cargo is a rectangular parallel pipe with dimensions of 7 × 2 × 1.5 m, giving a volume of 21 m^3;
(b) The material density is 2.5, giving a mass of about 50 t;
(c) The thickness of the steel walls of the truck is 0.5 cm;
(d) The steel density is 7.86;
(e) The driver is positioned 1.5 m from the cargo, on the cargo axis.

Loading of a truck

The worker in charge of the loading and unloading operations is assumed to be exposed to both the silo and the truck cargo. The scenario considered for the exposure to the truck cargo is similar to the scenario for the truck driver except for the position of the worker. The scenario considered for the exposure to the silo is the following:

(1) The silo is a cylinder 15 m high with a radius of 5 m;
(2) The material density is 2.5;

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[1] One becquerel of natural uranium is usually defined as 0.487 Bq of ^{238}U, 0.0227 Bq of ^{235}U and 0.487 Bq of ^{234}U . One becquerel of natural thorium is usually defined as 0.5 Bq of ^{232}Th and 0.5 Bq of ^{228}Th.

(3) The silo wall thickness is 1 cm;
(4) The silo wall is made from steel with a density of 7.86;
(5) The worker is located 5 m below the silo and at 5 m from the silo axis.

Employee in charge of handling bulk bags

The following scenario has been considered:

(i) The bulk bag is a 1 m cube;
(ii) The material density is 2.5;
(iii) The worker is positioned 0.5 m from the bulk bag.

Worker walking on a pile

The following scenario has been considered:

— The pile is a rectangular parallel pipe of dimensions 100 × 100 × 5 m;
— The pile density is 2.5;
— The worker is located on the pile, at 1 m above.

Dose rates due to external exposure

The external dose rates per unit activity concentration predicted from the calculations are shown in Table 2.

Influence of density

A density of 2.5 has been retained for the various scenarios. Calculations were repeated for material with densities ranging from 1 to 7. Similar results were obtained for the whole range of densities, with the dose rate varying by a factor of no greater than 2. Therefore a density of 2.5 continues to be assumed in the following sections, regardless of the type of NORM involved.

TABLE 2. DOSE RATE DUE TO EXTERNAL EXPOSURE

	Dose rate (μSv/h)			
	Truck driver	Loading of a truck	Handling of a bulk bag	Walking on a pile
Uranium	0.062	0.19	0.13	0.46
Thorium	0.092	0.29	0.20	0.69
Potassium	0.0058	0.18	0.012	0.04

TABLE 3. DOSE RATE DUE TO INTERNAL EXPOSURE

	Dose rate (µSv/h)	
	Inhalation	Ingestion
Uranium	4.8×10^{-2}	1.3×10^{-3}
Thorium	5.8×10^{-2}	8.7×10^{-4}
Potassium[a]	3.6×10^{-6}	6.2×10^{-6}

[a] In the human body, the potassium concentration is controlled so that, regardless of the amount incorporated, the body content of ^{40}K remains at about 0.060 Bq/g, giving rise to an effective dose of 180 µSv/a. Accordingly, ^{40}K contained in NORM is unlikely to increase the internal exposure.

3.1.2. Internal exposure

The internal dose rates for dust inhalation and ingestion predicted from the calculations are shown inTable 3. The following assumptions were made:

(a) A dust concentration of 1 mg/m^3;
(b) A dust ingestion rate of 1 mg/h;
(c) A breathing rate of 1.2 m^3/h;
(d) An inhaled particle size of 5 µm;
(e) The dust comprises particles coming from material with a parent radionuclide activity concentration of 1 Bq/g;
(f) For uranium and thorium, the lung clearance type is Type S [4];
(g) For the other elements, the lung clearance type is as specified in Ref. [4] for ‘all compounds’ or ‘unspecified compounds’.

3.2. Effective doses for realistic exposure scenarios

3.2.1. Determination of realistic scenarios

Two realistic exposure scenarios where considered:

(a) A truck driver;
(b) A worker in charge of handling bulk bags.

The external exposure scenario for the truck driver is as follows:

(1) The truck loading, transport and truck unloading (with the same exposure of the truck driver as during loading) each take 0.5 h;

(2) The driver makes 3 trips per day, giving a daily exposure period of 4.5 h;
(3) The driver works for 200 d/a, giving an annual exposure period of 900 h.

The external exposure scenario for the worker in charge of handling bulk bags is as follows:

(i) The worker is exposed for 4 h/d;
(ii) The worker works for 200 d/a, giving an annual exposure period of 800 h.

The internal exposure scenario for workers in dusty areas entails the same assumptions as those set out in para. 3.1.2.

3.2.2. Annual effective dose due to external exposure

Material intended to be processed for the use of its radionuclides

Figure 1 shows, for each radionuclide or material, the annual effective dose received by the truck driver and the bulk bag worker from external exposure in the realistic scenario described in Section 3.2.1, when the material is intended to be processed for the use of its radionuclides. The doses are calculated assuming that the activity concentration of each radionuclide is at the relevant exemption level defined in the Transport Regulations [1]. The results are in agreement with those of the other studies forming part of the coordinated research programme (especially those from Canada and Australia), with those reported in Ref. [5] and with the measurements provided in the industry studies used in this report.

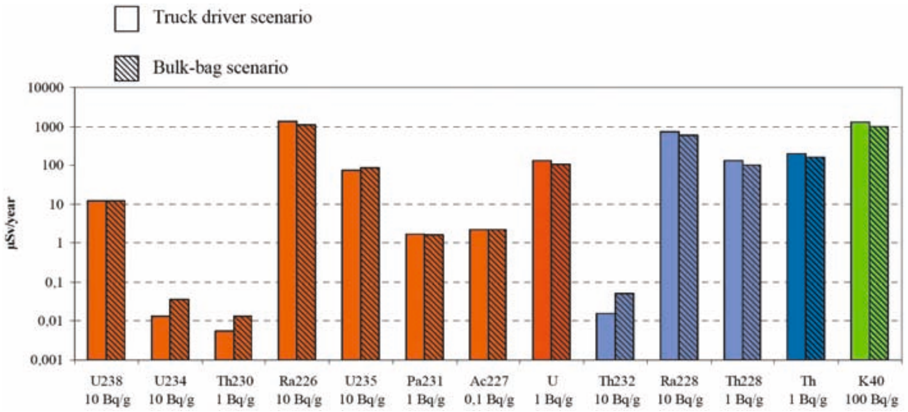

FIG.1. Annual effective dose due to external exposure for each radionuclide at an activity concentration corresponding to its transport exemption value.

Material not intended to be processed for the use of its radionuclides

The activity concentration criteria defining the scope of application of the Transport Regulations are 10 times higher than the exemption values. Thus, the annual effective doses due to external exposure received by the truck driver and worker handling a bulk bag in a realistic scenario, when the material is not intended to be processed for the use of its radionuclides, are 10 times those shown in Fig. 1.

3.2.3. Hourly effective dose due to internal exposure

Material intended to be processed for the use of its radionuclides

Fig. 2 shows, for each radionuclide, the hourly committed effective dose due to internal exposure for workers in dusty areas when the material is intended to be processed for the use of its radionuclides. The doses are calculated assuming that the radionuclide activity concentration is at the relevant exemption level defined in the Transport Regulations. It can be noted that this kind of exposure is not likely to be of concern to truck drivers, whose main type of exposure is external.

The activity concentration criteria defining the scope of application of the Transport Regulations are 10 times higher than the exemption values. Thus, the hourly doses due to external exposure received by workers in dusty areas, when the material is not intended to be processed for the use of its radionuclides, are 10 times those shown in Fig. 2.

4. CONCLUSIONS

NORM meeting the specifications given in para. 107 (e) of the Transport Regulations is outside the scope of application of those regulations. Where this is

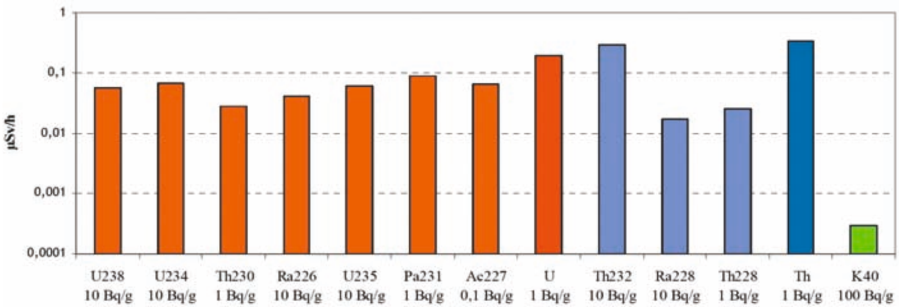

FIG. 2. Hourly effective dose due to internal exposure for each radionuclide at an activity concentration corresponding to its transport exemption value. Material not intended to be processed for the use of its radionuclides.

not the case, the transport of NORM has to be regulated if the activity concentration exceeds the exemption levels given in the Transport Regulations. This has two main implications in the light of the results reported here:

(1) The exemption levels are based on annual doses that are low enough to be considered reasonable for purposes of exemption. IRSN has assessed the doses received by workers (whether defined as ‘members of the public’ or not) in realistic exposure scenarios. The results of the assessment agree with those obtained in other studies and imply that any increase in the exemption levels for the transport of NORM would result in some workers receiving doses that might be considered unreasonable in the absence of any radiological monitoring. It is emphasised that doses received by such workers in a year may be significant.

(2) The current criterion for determining the scope of application of the Transport Regulations to NORM, namely a value of 10 times the exemption level, applies only to NORM not intended to be processed for the use of its radionuclides. This limitation appears to be consistent with the variability of concentrations of radionuclides in such material and the associated doses received by exposed workers.

Discussions are in progress at the international level concerning the possibility of removing the limitation described in (ii) above (especially in the case of uranium ore destined to be used in the nuclear fuel cycle). On the basis of the results reported here, it is argued that the exemption values in the current Transport Regulations should be retained.

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RAPPORTEUR SUMMARY OF TOPICAL SESSIONS 2 AND 3

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1. INTRODUCTION

This rapporteur summary covers the programme for the second day of the symposium, which included nine papers on the topic of regulatory aspects, three papers on the topic of transport of NORM and 10 posters on a variety of topics.

2. REGULATORY APPROACHES IN VARIOUS COUNTRIES

Presentations were made on various aspects of the regulatory approaches and application of international standards in three industrialized countries: the United States of America, Australia and Sweden. The presentations addressed principles and guidance established in these countries for the control of exposure to NORM.

In the case of the USA, the paper was written with the objective of providing an overview of the complex jurisdictional issues that pervade the regulation of NORM in the US legal system at both federal and State levels. It was concluded that a uniform and harmonized regulatory system was still a long way from becoming a reality.

Australia, despite also having to deal with the complexities of a federal structure, appears to have been more successful in implementing an approach to NORM regulation that is harmonized not only within the country but also with international standards. Australia has had a long history of dealing with radiation protection issues in a wide variety of NORM industries and is using this experience to compile practical guidance material for the various industries of concern.

The presentation from Sweden described work being undertaken to develop a regulatory approach to the management of NORM residues. The need for pragmatism was highlighted in addressing issues of exemption and clearance of such residues.

3. INTERNATIONAL CRITERIA FOR REGULATION OF NORM

For radionuclides in the uranium and thorium decay series, the IAEA Safety Standards recommend an activity concentration of 1 Bq/g as the criterion for considering whether material containing these radionuclides (other than material used for building) needs to be regulated as a practice. As reported in the opening session of this symposium, the 1 Bq/g criterion has now been incorporated into the draft revision of the International Basic Safety Standards, IAEA Safety Series No. 115 (the BSS). Material (other than building material) with an activity concentration of 1 Bq/g and below is not expected to give rise to an annual dose of more than about 1 mSv , but there has been some discussion on whether this is the case in all situations, particularly where there is a possibility of groundwater contamination (for example, mine residue deposits). A paper from Canada presented the results of a recent investigation into this issue. Using evidence based input parameters, the annual dose received by a member of the public from a large mine residue deposit with an activity concentration of 1 Bq/g was calculated. The maximum dose was found to be about 0.2 mSv/a using conservative, but realistic, assumptions. As expected, the majority of the dose was received via the groundwater ingestion pathway.

4. TRANSPORT OF NORM

The transport of NORM currently falls outside the scope of application of the IAEA Transport Regulations if the radionuclide activity concentrations do not exceed 10 times the exemption levels and if the material is not used for the extraction of its radionuclides. The IAEA, having just concluded a coordinated research project (CRP) on the topic of NORM transport, was now able to report some preliminary conclusions on the validity of this ‘10 times’ approach. It is generally apparent that the factor of 10 does indeed ensure an acceptable level of safety, and that a higher factor might apply in specific situations. Nine countries participated in the CRP. A separate presentation from one of these countries (France) provided the opportunity to gain a more detailed appreciation of the type of work carried out.

The transport of NORM is now becoming an issue not just for safety, but also for security. Since most of the world’s trade involves container cargo shipped in and out of seaports, it is important to screen this cargo for radioactive material to prevent, among other things, the smuggling of nuclear material in container traffic. The port of Antwerp is one of many large container ports around the world which is now equipped with sophisticated detection equipment and systems. The large quantities of NORM shipped in containers and the fact that such NORM is readily picked up by the detection systems has prompted the development of a new methodology to obtain an in situ estimation of the activity concentration of NORM in containers at ports. The paper describes how the deployment of such a system at the Port of Antwerp is now being followed up by further work to adapt the system for use in industrial facilities, where NORM is handled in bulk bags as well as containers.

5. RADIOLOGICAL ASSESSMENT OF NORM INDUSTRIES

An overview of NORM emissions in the Netherlands over the last 20 years was provided. Doses received by members of the public, attributable to these emissions, have been estimated. The main sources of these emissions were an elemental phosphorus plant, iron and steel plants and a titanium dioxide pigment plant.

The current status of NORM industries in Japan has been surveyed and a guideline for NORM was issued as a voluntary regulation last year to ensure safety. Through the implementation of this guideline by the industry, a decrease in the exposure of workers is expected.

A model has been developed in Germany for the characterization of NORM formation in different industrial processes. The model is based on mass and activity balances in industrial processes. It characterizes the radionuclide enrichment of a residue in relation to the input materials according to three main parameters: the mass transfer factor, the activity transfer factor and the enrichment factor. This model creates the possibility of assessing the radiological properties of materials in individual facilities in situations where raw materials are changed or processes are modified. This modelling of NORM becomes particularly important if residues are subsequently used in other processes.

6. USE OF NORM IN AGRICULTURE

In Brazil, phosphogypsum is used as a source of calcium and sulphur in agriculture and as an amendment for acid soils with high levels of aluminium. The poster presentation on this topic addressed a cost–benefit analysis to evaluate the impact of this practice on corn cultivation in the southern region of Brazil.

Tunisia is the world’s fifth largest phosphate producer. The characterization of ^{226}Ra, ^{232}Th and ^{40}K in phosphate fertilizer produced and exported by Tunisia and the determination of the ‘radium equivalent’ were described in a poster presentation. Triple super-phosphate fertilizer (TSP) exhibited the highest concentrations of ^{226}Ra (0.1862 Bq/g) and ^{232}Th (0.0337 Bq/g). The maximum radium equivalent for Tunisian fertilizers was about 0.238 Bq/g.

A poster from Thailand described how the extensive use of fertilizers had resulted in an enhancement of natural radioactivity in the river sediments and river bank surface soils of the Chao Phraya River.

7. USE OF NORM IN BUILDING MATERIALS

Building materials are one of the major sources of indoor radioactivity, besides the subsoil emanation of radon isotopes. In the south-west coastal region of India, which is known to have high natural background radiation mainly due to thorium rich monazite sand, a survey of radiation exposure was carried in dwellings. The major radiation contributors were the subsoil emanation of radionuclides as well as the uranium, thorium and potassium content of the building materials (cement, bricks and tiles). The results obtained in this paper may be useful for the definition of radiation safety standards by the authorized organizations in India.

8. CONCLUSIONS

The range of NORM issues and the ways in which they are handled can vary significantly between countries, as was evidenced by the various characterization studies presented. Furthermore, residues of large volume and low activity concentration may be considered for use as co-products, as evidenced already by the safe use of phosphogypsum as an agricultural amendment, as a treatment for sodic soils and as a construction material.

The IAEA provides global guidance on the appropriate levels of regulation of NORM industries and their residues (including the option of not regulating). However, there is an urgent necessity for a more **uniform and harmonized** regulatory scheme to offer guidance to those engaged in the generation, use, transport and disposal of NORM worldwide. Rephrasing the ALARA principle, I would like to invite you to think about a new variation: the AHARA principle, that is, ‘As Harmonized As Reasonably Achievable’.

MANAGEMENT OF NORM RESIDUES

(Topical Session 4)

Chairpersons

A. JANSSENS
European Commission

M. BELGHITI ALAOUI
Morocco

Invited Paper

A GLOBAL OVERVIEW OF NORM RESIDUE REMEDIATION AND GOOD PRACTICE

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Abstract

The issues of management and disposal of NORM residues have been a source of concern for many years in almost every country of the world. However, not all NORM industries have the same ideas when it comes to managing residues and their remediation. The volumes and types of materials vary considerably and so good practice is a situation specific activity, although the basic rules may be similar and the safety standards applied are the same. The phosphate industry has long been a major player in the issue of best practice management and disposal of NORM residues, but it is not always appreciated how the scale of the problems can vary. Similarly, with uranium production, not all the NORM residue issues are on the massive scale that is usually associated with mining operations. Also the oil and gas industries have been managing NORM residues for a long time but good practice is also a dynamic concept and another issue is to determine what is considered to be leading practice nowadays. It should also be pointed out that several other industries may be associated with NORM residues apart from these well known examples. The paper looks at a selected number of international sites and situations to show the variation of size and scope of problems both within and between industries and how good practice is developing in the areas of management, disposal and remediation of NORM residues at the affected sites.

1. INTRODUCTION

Materials containing naturally occurring radioactive material (NORM) are often processed for uses that bear no relation to their radioactive character. However, in many cases the residues from these processes contain radionuclides which have little or no commercial interest, yet pose issues of radiation safety and require protection measures to be considered not only during mining and processing of the original material but also when planning residue storage and/or eventual disposal The issue can be associated with a wide range of materials ranging from uranium and other metallic ores and mineral sands to phosphates, oil and gas and rare earth deposits; even the residues from water treatment plants may contain natural radionuclides in sufficient quantities to require specific management measures to be employed.

NORM related industries around the world are generally expanding as the consumer markets, and demand generally continues to increase yearly. Production of oil and gas and other energy related materials is growing steadily as societies and consumers demand more energy to support improvements in lifestyle and living conditions; metals are in ever increasing demand to provide the raw materials for the manufacture of consumer goods and expansion of new technologies; fertilizer production is foreshadowed to expand many-fold as demands for improved rates of food production are made by both developing and developed nations. All of these activities will result in increasing amounts of NORM residues that will require management and eventually disposal in one form or another.

Another significant matter is: Just which are the major NORM related industries? In 2006 the IAEA published a Safety Report on assessing the need for radiation protection in mineral related industries [1]. From this publication and other sources, the following ‘top 12’ NORM related industries may be identified:

(a) Uranium mining and processing;
(b) Rare earths extraction;
(c) Thorium extraction and use;
(d) Niobium extraction;
(e) Non-uranium mining, including consideration of exposure to radon;
(f) Oil and gas;
(g) Titanium dioxide;
(h) Phosphates;
(i) Zircon and zirconia;
(j) Metals production (Sn, Cu, Al, Fe, Zn, Pb);
(k) Burning of coal, lignite etc.;
(l) Water treatment, including consideration of exposure to radon.

A number of the industries listed are not usually associated with the need for radioactive waste management, which is why the introduction of appropriate good practices in the management of their residues and waste are so important. All of these types of operations should be screened to ascertain if indeed there are NORM issues relating to their residues and wastes. In many cases the outcome of the screening could be expected to show that there are no radiological safety issues and that all that is required is a good standard of ‘industrial housekeeping’ when it comes to residue management and disposal.

2. NORM RESIDUES AND REMEDIATION

As can be realized from the list of industries in Section 1, the range of materials that may need to be included when considering issues of management for NORM residues is very diverse. Apart from the more obvious residues from mineral processing, such as tailings, low grade minerals, contaminated process water, sludges, sediments and precipitates, contaminated plant and equipment and scrap metal, there may also be residues from the decommissioning and decontamination of processing and associated facilities and possibly gases and dust from flues at facilities where materials have been melted or heat treated in other ways. These in turn include scales and sludges removed from pipes and vessels, contaminated water from cleaning or decommissioning works containing dissolved and/or suspended contaminants, other liquids such as solvents containing dissolved and/or suspended materials, filters (in the form of cartridges, cloths, absorbents or other media) from air and water cleaning stages of decontamination work, and slag from any melting processes employed.

One of the common characteristics of many NORM residues is that whilst the specific activities of the material are low the volumes of material to be managed and remediated are very large. The classic examples include phosphogypsum from fertilizer production, residues from mineral sand mining, and even uranium mill tailings. However, there are also other NORM residues which are relatively small in volume but which can have very high activity concentrations; the scales derived from production waters in the oil and gas industry are a typical example of such material. Each of these residue materials has to be disposed of in an appropriate manner to ensure the long term safety of the population and the environment.

The major issue for the safe management and remediation of these residues is to ensure that, before disposal is determined as the only option, all opportunities for use/reuse, volume reduction and recycling of the residues have been evaluated and that the final remediation and disposal solution is demonstrably an optimized solution. The selection of processing options that will optimize financial returns as well as minimizing wastes and maximizing opportunities to find other uses for residues is now one of the most important elements in the planning and development of every project that plans to exploit NORM.

In all NORM processing operations, every effort should be made to minimize the amount of residue generated, especially in the case of residues that may eventually be declared to be waste. There will always be situations where there are wastes requiring disposal despite all efforts to minimize waste having been employed.

The options for the disposal of NORM waste require that some basic criteria are met; these in turn may dictate that only a limited range of options is available. The main considerations in NORM waste disposal are:

(a) The waste should stay contained for as long a time as possible;
(b) The waste should be contained in such a way that discharges to the air and to surface water and groundwater resources are minimized to the greatest extent practicable;
(c) The waste disposal facility should be designed to be operated with the minimum maintenance requirement and minimum level of institutional control practicable for the long term future.

Given these criteria, the most frequently used options for NORM waste disposal are:

(1) Custom built containments such as above ground tailings dams;
(2) Dilution and dispersion into an approved receiving environment;
(3) Burial below ground level in custom built pits, natural caverns or old mine workings and similar facilities.

Generally, the dilute and disperse option has been the least acceptable to society. When one considers former practices of discharging large volumes of residues such as mill tailings or phosphogypsum into rivers, lakes or the ocean this is not surprising. However, in the mineral sands industry it is now an accepted practice to return radioactive waste into the remediated land form after incorporating it into the soil so that the level of radioactivity is no more than was found in the original ore — in effect returning the radiological condition of the remediated site to the pre-mining state.

The use of above ground waste storage is not seen as the best option but it may be the most appropriate practice in a site specific situation. Today, containment structures such as tailings dams are required to be built using appropriate standards, which was not always the case in former times. Modern tailings containments are often required to be built as if they were going to be water retaining structures. This has reduced the risk of failure of tailings containments and improved levels of safety for the environment and the downstream population.

The option to place wastes below ground is attractive as a long term solution but the location must be suitable in order to avoid setting up the potential for later (and possibly long term) contamination of water resources. Some technologies such as thickened tailings and paste tailings are being examined and selected for use in suitable locations. The reduction in volume and increased structural strength of such wastes placed using these technologies may increase stability and safety as well as allowing increased exploitation of the mineral resource in the workings.

But today many of the residue management options will begin with looking at ways to minimize waste and most frequently this may be achieved through finding alternative uses for all or some of the residues. The solutions are usually specific to an industry, a site, a source material or even a processing option. In the following sections these issues are examined for a number of established NORM industries and the successful remediation examples are also included.

3. NORM INDUSTRIES

This section considers in turn eight of the major industries associated with NORM residues. The range of industries has been selected to indicate the range of materials and volumes that may be associated with these remediation issues.

3.1. Phosphate industry and phosphogypsum

One of the commonly found, largest volume and most widely distributed NORM residues is phosphogypsum (PG). The quantities of material involved can be very large indeed as there may be up to 5 t of PG produced for each tonne of fertilizer product. In Florida it has been estimated there are 1 billion t of PG already in stacks and this total increases by about 30 million t each year; at Huelva in Spain there are 120 million t of PG stacked alongside the Rio Tinto river; there are over 390 million t of PG throughout Brazil associated with fertilizer production; and these volumes increase steadily as the world’s requirements for fertilizer increase year by year [2].

What options are available to producers for managing these residues and their eventual remediation? In the jurisdictions of some authorities, such as the United States Environment Protection Agency (USEPA), PG has been declared a hazardous waste and so disposal is the only option. In other areas, efforts are being made to discover possible options for other uses of the material. For example, in Jordan and Brazil work is under way to determine if PG can be safely used as a feedstock for the manufacture of construction blocks. At the IAEA, examination of data from around the world has shown that approximately 50% of the global PG stockpile has a radionuclide activity concentration of less than 1 Bq/g and is thus outside the scope of regulation as recommended in the IAEA’s own safety standards [3]. Subject to the PG meeting other safety criteria, especially with respect to potential issues of heavy metal or other chemical contamination, it is hoped that much of this class of PG may be used as a construction material as either building blocks, a cement extender or in dry cavity wall boards.

Other remediation and disposal options have included studies of using PG in the construction of all weather market and feeder roads in areas where other additives such as cement may be in short supply or too expensive. Such roads do not have very heavy traffic roads and the experience from more than 10 years of studies in Florida, for example, show that such roads are very durable and may require less maintenance than comparable, conventionally built roads [4]. There are also studies under way to examine the use of PG as an additive in municipal landfills. In these situations it appears that the residual phosphate offers an energy source to microbes that then digest various components of the landfill more efficiently and so reduce the overall volume of the mass of refuse.

In some locations, PG is being used as a soil conditioner that also offers a small nutritional benefit to crops such as ground nuts as a consequence of the residual sulphur, often an essential trace element in plant nutrition and present at low levels in PG.

There have also been cases where phosphogypsum disposal areas have been revegetated, such as at Heulva in Spain, and have become open space with the potential to develop as public recreation areas or nature conservation areas (Fig. 1). Provided a soil cover can be made available, the establishment of vegetation on

FIG. 1. Huelva, Spain, revegetated PG — note stork in nesting box on stand.

FIG. 2. NORM contaminated items in storage prior to cleaning or disposal.

such sites has not been difficult and whilst they are unlikely to ever be suitable for urban housing use they can become a significant urban asset in terms of green space.

3.2. Scrap metal

Another source of NORM residues is found in scrap metal associated with NORM processing industries. The scrap can include worn out or broken items of plant and equipment such as pumps, trucks and bucket conveyors, old processing equipment and pipe work (Fig. 2).

The most common forms of residue are process residues, scales and sludges that are deposited on the inside of pipework that has carried water from deep underground sources such as in the oil and gas industry or some water supply sites. These scales and sludges have various compositions and, depending on the exact composition, the concentrations of some radionuclides can be very significant. The hard scales are generally dominated by radium (^{226}Ra and ^{228}Ra) with concentrations of up to 2800 and 15 000 Bq/g respectively reported. In the sludges, the dominant radionuclide is often ^{210}Pb with concentrations of up to 1300 Bq/g reported [5]. Such residues may cause workers' external exposure to be increased due to gamma radiation during the operational part of a plant's life; but at decommissioning and dismantling the presence of such residue deposits may be very hazardous to the workers involved as they pull apart and even cut up items such as pipes and tanks, both in terms of external dose and internal dose if the workers are not properly equipped and supervised.

The disposal of these sludges and scales requires an approach which will minimize risk to the workers, environment and the community and ensure that the threat remains contained. Decontamination of NORM scales and sludges will also potentially generate further residues and waste as the cleaning media become contaminated and accumulate. Examples include grit and beads used in surface blasting, water from high pressure washing, solvents, strippable coverings and removable linings. The water may be evaporated and the evaporites then disposed of with the other wastes. Often, secure containment in appropriate containers and then secure, deep burial is the course of action taken. Systems which finely divide such high activity concentration materials for dilution into a low activity matrix are not an acceptable practice.

3.3. Oil and gas

The NORM residues associated with these industries are essentially similar to the scales and sludges described in Section 3.2. The scales are very common on pipework that has contained formation or production water brought to the surface in association with oil and/or gas. The major source of residues comes from the decontamination and cleaning of equipment such as pumps, pipes and holding tanks when they are being cleaned for reuse or disposal to another location or owner (that is, sold). The residues from the decontamination and cleaning activities cannot usually be reused or recycled and so the volume is minimized and then the final residue disposed of with the waste. Care should also be taken in the management of any evaporates generated from the formation water in areas where such water is allowed to flow to an area for surface disposal by irrigation and natural evaporation.

Where there are enclosed areas or vessels that have contained scale, radon monitoring should be undertaken as well to ensure that the potential for exposure of workers by all pathways is managed. This is especially important when vessels may take some time to inspect.

3.4. Mineral sands

In the mineral sands industry the NORM residue of concern is monazite, a mineral which contains thorium as the primary radioactive constituent. Although thorium was in demand in the past as a material for use in gas mantles, cinematographic lenses, welding rods and alloys it has been replaced by non-radioactive components in many cases. As a consequence, although monazite may be processed to remove rare earth elements which are not-radioactive, the bulk of the material that remains after these processes is a NORM residue for which there is often no significant market at this time. Thus it is regarded as a waste and requires disposal. In a number of locations the ‘dilute and disperse’ option has been used where it is easy and practical to do so. One example is in the mineral sands operations of Western Australia. There the monazite residues are returned to the original excavation site and blended and incorporated back into the landscape during the remediation earthworks at the site until the level of activity concentration is no more than it was in the original excavated material. Thus the natural background situation pre-mining is essentially reproduced and radiation levels are acceptable to the authorities. In Western Australia the main limitation on reuse of some sand mining areas relates to them still containing water resources used by the utility companies for potable water supply and the restrictions refer to possible chemical contamination as a consequence of human activity such as use of fertilizers. Elsewhere in the region some land has been restored for wildlife reserves and some to agriculture following remediation.

3.5. Uranium production

The uranium mining industry is often not included in consideration of NORM related issues and yet it is treated in safety terms in exactly the same way as other NORM industries. The most obvious residues from uranium mining are the tailings, ground up rock from which the uranium has been extracted; sub-economic mineralized material (sometimes referred to as BOGUM or proto-ore) which contains sufficient uranium for the material to be classified as radioactive material, but not enough to warrant the cost of extraction; process residues, laboratory wastes, scales and sludges from various parts of the extraction and concentration processes; contaminated scrap metal and machinery no longer needed in the operation (Fig. 3); and waste waters containing levels of natural radionuclides in excess of regulatory limits.

There have been many studies on the options for managing and disposing of tailings and the consensus is that disposal below ground level is best [6, 7] if suitable opportunities are available. These include using a mined out pit, backfilling stopes and similar workings in underground mines, or even placement in custom built silos excavated underground. Such situations all have to be assessed for suitability and safety, particularly with respect to the potential to contaminate underground water resources. Below grade disposal has the advantage that dispersal of the residues is unlikely to happen except within a geological time frame, and so the risks from contamination due to the long lived (but low specific activity) radionuclides are reduced considerably.

FIG. 3. Uranium process residues in scrap pipework.

The tailings may be dewatered through the use of paste technology prior to placement, thus reducing the volume of space required for disposal. Tailings can also be mixed with pozzolanic material such as fly ash or cement before placement, which will help solidify the mass and thus reduce further the risk of dispersal of contaminants. In some instances tailings have been disposed of jointly with waste rock in so-called ‘co-disposal’ schemes, again to try and improve the packing of the residues into a given volume and reduce the risks of dispersal of contaminants.

It must always be remembered that as with all mining residues the major risk may be from chemical and/or physical properties of the material and not only or primarily the radioactive nature of the material. Such residues must be fully characterized before disposal plans are finalized to ensure that all risks are being fully understood and appropriate management strategies are in place for all the identified risks and their possible consequences. Only then can all the parties involved be certain that the optimum protection is afforded to the population and the environment over the longest term practicable.

Where residues have to be disposed of above ground level, for instance in tailings dams or in waste rock piles, every effort should be made to ensure that the residues are fully characterized so that appropriate risk management strategies can be implemented. The radiological concerns are usually external gamma dose rate and possible radon exhalation. Both of these are generally controlled through the use of a cover system over the final landscape land form. Such cover systems often perform multiple functions: to contain the residues for an appropriate time; to restrict ingress of water which may leach out chemical contaminants; to prevent dispersal of residues through entry of burrowing animals or persons seeking to reclaim scrap materials; to shield against gamma radiation; and to restrict radon exhalation rates to levels that are deemed acceptable. All of these functions should be achieved in accordance with standards that are acceptable to the regulating authorities. Tailings dams above ground should be designed to ensure that the opportunities for seepage of any liquids, generated by or passing through the residues, to groundwater through the walls and floor of the facility are minimized, as well as achieving the necessary performance criteria for the cover system to manage the issues previously mentioned. Such seepage restrictions may be achieved through a combination of natural and/or synthetic liners and associated engineering controls such as seepage collector systems. The operational systems associated with these facilities should be such that the operations are effective through passive management to the greatest extent practicable. Also, the designs should meet any longevity requirements set by the regulatory authority with the need for maintenance made as minimal as possible.

In some cases, above ground disposal sites may be suitable for some further use. Examples of this are a golf course built over a remediated uranium mine waste rock dump in Germany [8] and a public park on a remediated former mill tailings site in Grand Junction, Colorado, after the relocation of the residues to the secure disposal cell at Cheyney, some 20 km outside the town.

3.6. Metal slags

When metals’ ores are processed by smelting to separate the desired (metal) fraction from the worthless fraction, much of the remaining material becomes a residue known as slag. This material is often hard, may be semi-vitrified and intractable after cooling and may be granular or massive in form. Slag is generally a mixture of metal oxides, although sulphides may also be present if they were found in the original ore. Previously the main use for slag was as a granular fill material (Fig. 4), although the basic slag from steel making was often used as a soil amendment since it had some neutralizing capacity and could also release small amounts of phosphorus into the soil. More recently such slag materials have been investigated for use as additives to cement where they can improve resistance of concrete to attack by chlorides and sulphates. Whilst NORM is not generally an issue in ferrous smelting (iron) the slag from smelting metals such as copper and lead may contain sufficient concentrations of radionuclides to attract the attention of regulating authorities, especially in the case of copper minerals that may contain significant amounts of uranium.

Once the activity concentration of the slag and the homogeneity of the material have been established, a plan for disposal for alternative use of the material can be formulated as appropriate. Again, care must be taken to check

FIG. 4. Artisanal copper smelter; at right, slag spread as fill for future building foundations.

that there are no other significant environmental or health risks in the proposed use of the material due to its other chemical and/or physical properties.

Care must be taken at workplaces where smelting is done to ensure that there is no escape of NORM residues in the gases and dust that may be generated by the process. Air scrubbers, dust traps, baghouses and precipitators should all be employed as required to restrict the loss of residues to the atmosphere. Again, the material trapped by these systems will itself require management as a NORM residue, although in most cases the material is likely to be declared waste and sent directly to disposal; the same would apply to residues remaining after the cleaning and decontamination of equipment associated with these systems.

3.7. Water treatment

Water treatment works may be a source of NORM residues, especially where the water being treated may be from deep geological formations and contain enhanced levels of radium and uranium. The residues from water treatment may be in a variety of forms. Radioactive scales containing radium and other radionuclides may accumulate in pipes, similar to the way they are found in the oil and gas industry. The disposal of these pipes as scrap metal or the disposal of residues generated after the cleaning of such pipes do present a waste management issue. The best solution is often to concentrate the radioactive material and contain it for disposal in an appropriate manner such as a deep disposal cell or borehole.

Water that comes from deep underground is usually quite warm and so often is passed through cooling towers to cool it before treatment to become potable water. The walls and baffles in such cooling facilities often become encrusted with scales and similar salt deposits left by the water as it cools and evaporates. When these areas are cleaned the residues should be regarded as NORM and appropriate workplace safety practices employed as necessary. Also as it reaches the surface such water may degas and thus there is a potential for increased levels of radon to be present in the immediate vicinity of degassing areas when compared to the general background levels. This is especially true in water treatment facilities that may be located underground or otherwise poorly ventilated.

The actions of purifying and cleaning water may also generate NORM residues, for example on filter cartridges, ion exchange resins and reverse osmosis membranes and similar locations where radionuclides may accumulate; eventually the spent filters require disposal. Precipitation of metals by addition of liming agents results in the possible accumulation of radionuclides and heavy metals in the precipitate. Again, each site will need to be characterized to establish the level of risk and whether the material falls within the scope of regulation of radioactive material in the local jurisdiction. Such precipitates may be managed in a similar way to PG residues, for instance by disposing of in landfill facilities or diluted into soil as a conditioner, depending on the characteristics of the specific material. Again the non-radioactive content of the material must also be characterized. In some cases the presence of metals could be at levels sufficient to preclude any further use or options other than disposal to a secure landfill or disposal cell.

A further example of a NORM residue from water treatment may be found in some parts of the former Soviet Union. Formation water from oil and gas deposits was passed through finely divided coal which acted as active carbon to sequester elements dissolved in the water, in particular iodine and bromine, but also radionuclides. The iodine and bromine were recovered for commercial use whilst the now radioactive coal dust was discarded as a residue. The residue was rarely disposed of in a safe manner and, after the sites were abandoned when the Soviet Union was dissolved, much of the residue was stolen from the abandoned sites and used as fuel for brickworks and in homes. The pipes which contained the water also accumulated radium scale and so they too became a residue of concern (Fig. 5). It is hoped that various remediation programmes currently being planned will enable these residues to be collected up and disposed of in secure containments as no further beneficial use has been identified for them.

FIG. 5. Asbestos cement water pipe with radium scale and radioactive coal dust residues.

3.8. Coal and fly ash

Coal deposits have a very variable content of uranium and other radionuclides and at various times in the past efforts were made to recover uranium from coal or from the ash. Today the major source of coal ash is the flyash which is produced as the residue from modern coal fired power plants. There are also bottom ash and boiler slag which are additional residues of coal burning. The volumes of material produced are again very large, up to 100 million t per year in the USA and 10 million t per year in the United Kingdom alone. Global production may be as more than 400 million t. However, the flyash is rarely considered NORM as it usually has an activity concentration below 1 Bq/g and is thus not truly NORM since it is not subject to regulation for radiological reasons. The most common use for flyash is as an extender in cement and the manufacture of building blocks. In the USA it has been estimated that 45% of the ash is recycled in this way [9]. The dilution of the material in the final product means that the radioactivity levels are rarely of any concern in a regulatory context. The other ash materials from coal burning may also be used in similar ways.

4. DISCUSSION AND CONCLUSIONS

From the previous paragraphs it can be seen that many NORM related industries produce residues that become of concern when the concentrations of radionuclides exceed the threshold of the regulatory limit for definition of radioactive material. Some of these residues, particularly those that are of large volume and low specific activity, may be considered for use in a number of applications including use as construction material in roads, building blocks, cements and dry walling boards; as an agricultural soil amendment; and as a landfill additive. Alternatively, they may be disposed of by dilution back into the environment under appropriate and suitable circumstances.

On the other hand, some NORM residues are characterized by being of high specific activity but relatively small volume; for these residues immediate disposal into long term containment is usually the only option. There are also issues of managing the disposal of residues such as pipework and machinery (both mobile and static) that may have been contaminated by NORM residues. In some instances, cleaning and decontamination of such items may be economically feasible — but then there is the issue of managing the residues from the cleaning and decontamination processes which are a new NORM residue. Again, the activity concentration and volume of the residues will be the factors that determine the optimum disposal strategy.

When dealing with NORM residues care must be taken to avoid forgetting that other properties of the residues, especially chemical properties, may be more limiting on disposal options than the radionuclides.

The range of material types, forms and levels of specific activity that may be encountered when dealing with NORM residues is extensive. As NORM issues are brought more into the public domain and more industries are identified as having potential to produce NORM residues the question of appropriate, safe and secure residue disposal becomes more and more significant.

A pragmatic, risk-based approach to safety should be adopted when examining disposal options for NORM residues. The energy that has been used to produce such residues should not be wasted by immediately assuming that disposal is the only option. Hopefully this will improve the efficiency of many industrial processes and reduce the amount of NORM residues being set aside as waste when they may have value as feedstock to another process or for reuse in another industry. There are already several options for further use or recycling of many of these residues and research is under way to determine what other options may become available in the future. Looking at the stacks of PG in Florida one is struck by the notion that there has to be a better way of managing those particular residues; and one that has a better long term outlook.

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Invited Paper

DECOMMISSIONING OF A RARE EARTHS EXTRACTION FACILITY

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Abstract

The occupational and environmental radiation exposures and radiation safety issues associated with the decommissioning of a rare earths and thorium extraction facility are presented. This plant was in continuous operation during the period 1952–1988, chemically processing monazite for the separation of rare earths and thorium. It was decommissioned over a nine month period during the early 1990s. Operations at the plant over the years had resulted in buildup of activity on equipment surfaces, floors, pipes, walls and associated structures. The reaction tanks had accumulated elevated levels of activity as a result of impregnation of radionuclides into the rubber linings and pitting of surfaces due to chemical corrosion. Gamma exposure levels, airborne radioactivity and concentration of radionuclides in sludge and wastes for disposal are presented. The sequence of systematic decommissioning operations is outlined. The solid, liquid and gaseous effluents generated are characterized and quantified. Personal radiation exposures for the decommissioning and waste disposal operations are estimated. Novel methods of decontamination and recycling of large metallic objects and building surfaces are presented as a means of optimizing the quantity of waste as well as radiation exposures. The total effective doses received by the workers from external and internal exposures over the duration of the decommissioning operations were assessed to be in the range of 0.20–8.94 mSv, with a mean value of 7.2 mSv. After completion of the decommissioning operations, the groundwater was monitored for a number of years to check for contamination from the waste disposal sites. The ^{228}Ra concentrations in the groundwater were very low.

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1. INTRODUCTION

The plant engaged in the processing of monazite for separation of thorium and rare earths in India was in continuous operation for nearly four decades. It was decommissioned mainly due to structural weakness and corrosion. The decommissioning process covered a period of nine months during 1990–1991. The major process steps involved in the plant were caustic soda digestion of the mineral, filtration of mixed thorium and rare earths hydroxide, extraction of rare earths from thorium, filtration of thorium hydroxide concentrate and deactivation and casting of rare earths chloride. Except for the caustic soda digestion unit, all downstream process equipment and buildings were identified for decommissioning. The paper gives the details of gamma dose rates, contamination status, individual exposures and the disposal of radioactive waste involved in the decommissioning operations.

2. RESIDUAL RADIATION AND CONTAMINATION LEVELS

Operations at the plant over the years had resulted in a buildup of activity on equipment surfaces, floors, pipes, walls and associated structures. The reaction tanks had accumulated elevated levels of activity as a result of impregnation of radionuclides into the rubber linings and pitting of surfaces due to chemical corrosion. This was observed particularly in the tanks used for deactivation of the rare earth chloride, with the absorbed dose rate on the internal surfaces of the tanks having risen from less than 200 µGy/h in 1970 to nearly 1200 µGy/h in 1989. The activity was associated mainly with ^{228}Ra and its progeny. The activity concentration of ^{228}Ra in the rubber linings was 55 500 Bq/g, considerably higher than the value of 13 000 for the $Ba(Ra)SO_4$ produced in the deactivation process. The gamma dose rates at the time of decommissioning are shown in Table 1.

Alpha contamination levels on floors and equipment surfaces varied between 1.5 and 333 Bq/cm^2. The walls showed appreciable levels of contamination only up to a height of 3 m. The activity concentrations of airborne dust and thoron progeny prior to decommissioning are shown in Table 2. The average thoron progeny concentration in the thorium filter press area increased from about 6 µJ/m^3 in 1983 to about 37 µJ/m^3 in 1988.

TABLE 1. EXTERNAL EXPOSURES IN THE RARE EARTHS PLANT PRIOR TO DECOMMISSIONING

	Absorbed dose rate (μGy/h)	
	General working area	On equipment
Entrance	50	—
Deactivation tanks	800–1200	1000–2500 (outside) Up to 5000 (inside)
Extraction tanks	50	30–60
Weak chloride tanks	—	100
Wooden tanks	50–100	200–250
Central passage	300–500	—
Thorium slurry tanks	60	150
Extraction platform	30	—
Deactivation platform	800–1000	—
Thorium press	10–40 (floor)	20–50
Mixed cake ($PbS+Ba(Ra)SO_4$) press	30–200	100–400
Slurry pipes	—	5–100
Switch panels	—	5–10
Motors and pumps	—	5–600
Moore filters	30–70	30–60

TABLE 2. DUST AND THORON PROGENY EXPOSURE LEVELS IN THE PLANT PRIOR TO DECOMMISSIONING

	^{232}Th activity concentration in airborne dust (mBq/m^3)	Potential alpha energy concentration of thoron progeny (μJ/m^3)
Thorium concentrate press floor	34	36.7
Mixed cake press floor	12	26.7
Deactivation section	15	9.0
Thorium concentrate drying area	58	13.5
Mixed cake cask filling area	52	6.2
Extraction section	12	5.2
Wooden tanks area	24	24.8
Washing machine	8	58.3
Moore filters	23	4.2

3. DECOMMISSIONING OPERATIONS AND WASTE DISPOSAL

The sequence of decommissioning operations was as follows:

(a) Sludge was removed from pipes, vessels, pits and drains by pumping or by manual recovery into polyethylene lined concrete casks in readiness for disposal.

The rubber linings of the extraction and deactivation tanks were not amenable to decontamination. Instead, the tanks were cut open from outside by gas cutting torches and the linings were removed using scraping tools with long handles. This avoided workers having to enter the tanks where the gamma exposure levels were very high. The lining fragments were transferred to concrete casks for disposal. The sections of the tanks and other contaminated metallic items were decontaminated to the extent possible by garnet sand blasting in ventilated booths with exhaust air scrubbing. The workers engaged in theseactivities were equipped with full protective clothing and compressed air line respirators and were given suitable instructions and training.

(b) Items with fixed contamination such as wooden tanks, panels and frames of filter presses, wooden platforms, Moore filter assemblies, agitators and supporting structures and pipes were, as necessary, dismantled, cut into small pieces and cleaned to remove any loose surface contamination prior to disposal.

(c) Pumps, electric motors and other electrical equipment were decontaminated and salvaged for reuse after having been checked for residual contamination.

(d) Contaminated floors and walls were scrubbed and washed to remove loose contamination. The remaining fixed contamination was then removed by chipping away the plaster layer. The contaminated chippings were collected in HDPE bags for disposal.

(e) The buildings, after the removal of equipment and contaminated floor and wall surfaces, were demolished. The structural components and rubble were salvaged or disposed of as non-radioactive waste.

The disposal methods for the various wastes are summarized in Table 3. The experience gained in the decommissioning operation demonstrated that the amounts of contaminated material requiring to be disposed of as waste could be significantly and cost effectively reduced as a result of decontamination of the building structures by removing the plaster layers from floors and walls and decontamination of metal surfaces by sand blasting. In the case of metallic items, the volume of waste was reduced by a factor of 10.

TABLE 3. DISPOSAL OF DECOMMISSIONING WASTES

	Volume (m^3)	Absorbed dose rate (µGy/h)	Disposal method
Sludge in concrete casks	20	50–500	Concrete trenches
Liquid residues from tanks and decontamination operations	300	—	Treatment at effluent treatment plant followed by discharge
Rubber tank linings in concrete casks	10	50–5000	Concrete casks in concrete trenches
Mild steel items not fully decontaminated	25	1–200	Concrete trenches
Wooden items	27	30–200	Concrete trenches
Plastic items not fully decontaminated	3–6	1–50	Concrete trenches
Wall and floor chippings in HDPE bags	15	1–10	Between casks in concrete trenches
Contaminated garnet from sand blasting	—	—	Concrete casks in concrete trenches

Exposure levels for airborne dust and thoron progeny during the work are shown in Table 4. The total effective doses received by the workers from external and internal exposures over the duration of the decommissioning operations were assessed to be in the range of 0.20–8.94 mSv, with a mean value of 7.2 mSv. After completion of the decommissioning operations, the groundwater was monitored for a number of years to check for contamination from the waste disposal sites. The ^{228}Ra concentrations in the groundwater were very low (<0.001–0.004 Bq/L).

4. CONCLUSIONS

The decommissioning operations of the rare earths extraction facility were carried out systematically and 40 personnel were engaged for the operation resulting in a collective dose of 288 man·mSv. The individual exposure was 0.2–8.9 mSv with an average of 7.2 mSv. The entire operation involved disposal of 340 GBq of active waste of which 99% was contributed by solid wastes. The decontamination of building structures by removing plaster on the floors and walls has proved to be a good method for reducting quantities of low level wastes to be handled in addition saving the space for disposal. Decontamination of metallic surfaces by garnet sand blasting under controlled conditions has been

TABLE 4. EXPOSURE TO AIRBORNE RADIOACTIVITY DURING DECOMMISSIONING OPERATIONS

	Airborne dust		Thoron progeny potential alpha energy concentration (μJ/m^3)
	Mass concentration (mg/m^3)	^{232}Th activity concentration (mBq/m^3)	
Sludge removal	0.10	10	6–17
Dismantling of services	0.10	10	6–17
Dismantling of tanks	0.19	^{228}Ra: 40–60 (mean 50)	6–8
Gas torch cutting of tanks and removal of rubber lining	0.33	^{228}Ra: 70–2720 (mean 900)	6–8
Scrubbing and washing of floors and walls	0.20	10	6
Chipping of floors and walls	0.54	30	2–4
Sand blasting of contaminated items[a]	120–180	230–650 (mean 440)	0.6–2
Demolition of buildings	0.2–0.5	10	0.6–2

[a] The actual exposures of workers are much lower than the levels quoted because of the use of air line respiratory equipment.

demonstrated as a cost effective means of decontamination and disposal. This method resulted in significant volume reduction by a factor of 10 of the active wastes. Some of the decontaminated metals also have reusable values.

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A STUDY ON EVALUATION METHODS FOR ^{210}Pb ACCUMULATION ON ADSORBENTS OF NATURAL GAS TREATMENT PROCESSES

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Abstract

Evaluation methods for ^{210}Pb accumulation on the adsorbent of natural gas treatment processes have been developed. These evaluation methods enabled not only the prediction of the ^{210}Pb concentration in the spent adsorbent but also the determination of the change intervals for the adsorbent in order to keep the ^{210}Pb concentration in the adsorbent below the exemption level. For the prediction of the accumulation of radon decay products, two methods have been developed for evaluating ^{210}Pb accumulation in the adsorbent used in the natural gas treatment process. The 'principle method' evaluates the ^{210}Pb accumulation based on the radon absorption equilibrium coefficient of the adsorbent. The coefficient is determined by a breakthrough curve obtained by an adsorbent column test using surrogate gas including radon. The 'practical method' evaluates the ^{210}Pb accumulation based on radon adsorption determined by column tests using real process gas. The laboratory tests indicated that the practical method is an easy and practical evaluation method.

1. INTRODUCTION

Natural gas (NG) includes the natural radioactive gas radon (^{222}Rn), which is a decay product of ^{226}Ra existing in the earth's crust. It has been reported that the range of radon concentrations in NG ranges from zero to 200 000 Bq/m^3 [1].

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NG is treated using adsorbents for removing impurities such as water, carbon dioxide and sulphur compounds in NG processing plants and LNG plants. The decay products of radon such as ^{210}Pb, ^{210}Bi and ^{210}Po may accumulate on these adsorbents because radon is adsorbed on the adsorbent together with impurities. The half-life of radon is 3.8 d and the decay products of radon remain on the adsorbent. Therefore, ^{210}Pb, which has a half-life of 22.3 a, accumulates until the adsorbent is changed. It has also been reported that the radioactivity concentration of spent adsorbents exceeded the exemption level and the spent adsorbents were disposed of as radioactive waste in the case of the Jumping Pound gas plant in Canada [2].

It is necessary to determine whether the radioactivity concentration of spent adsorbent is above the exemption level or the unrestricted release limit, where the exemption level of ^{210}Pb is specified as 1 Bq/g by the IAEA [3], 0.2 Bq/g by the International Association of Oil and Gas Producers [4] and 0.3 Bq/g by Health Canada [5][2], among others. However, it is difficult to measure ^{210}Pb and its decay products ^{210}Bi and ^{210}Po in a vessel from the outside during operation because these radionuclides do not emit high energy gamma rays. As a result, the characterization of the spent adsorbent has been carried out after a change of adsorbent using radiochemical analysis. Therefore, evaluation methods for ^{210}Pb accumulation before such change are needed. Two kinds of evaluation methods for ^{210}Pb accumulation on the adsorbent materials were studied in this paper.

2. THEORY OF ESTIMATION FOR ^{210}Pb ACCUMULATION ON ADSORBENT MATERIALS

The estimation methods of ^{210}Pb accumulation on the adsorbent materials were developed using the following assumptions:

(a) The mechanism of radon adsorption is physical adsorption and the amount of radon in the adsorbent is proportional to the radon concentration in the gas;
(b) The concentration of radon in NG and the amount of radon adsorbed in the adsorbent is constant during the operating period;
(c) Decay products of radon are retained on the adsorbent material;
(d) The decay of radon produces ^{210}Pb directly, because the half-lives of the short lived radon decay products ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po are negligibly short when compared with that of ^{210}Pb;

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[2] Health Canada provides this value as the 'unconditional derived release limit for diffuse NORM'.

(e) During operation and regeneration, radon decay products are retained on the adsorbents, and the regeneration period can be corrected by multiplying the operation rate τ by the amount of radon adsorbed, where τ is the ratio of operation days of the adsorbent to the change interval.

Based on the above assumptions, the rate of change of $n_{Pb\text{-}210}$ (the concentration of ^{210}Pb atoms, in atoms/g) was described as:

$$\frac{dn_{Pb210}}{dt} = A_{Rn} - \quad {}_{Pb210} \cdot n_{Pb210} \tag{1}$$

where

λ_{Pb210} is the decay constant of ^{210}Pb (s^{-1}),
A_{Rn} is amount of radon adsorbed on the adsorbent (Bq/g),

and A_{Rn} is calculated using the equation:

$$A_{Rn} = \frac{P}{P_a} k_{i\,Rn} \cdot C_{Rn} \tag{2}$$

where

P_{Pb210} is the treatment pressure in the adsorption process (MPa)
P_a is the atmosphere pressure (0.1 MPa)
$k_{i\,Rn}$ is the adsorption equilibrium coefficient of radon in the adsorbent i (m^3/kg),
C_{Rn} is the radon concentration in NG (Bq/m^3).

The ^{210}Pb concentration on the adsorbent A_{Pb210} (Bq/g) at the end of the operating period T (s) is obtained by solving the differential equation (1) based on the initial condition $n_{Pb210} = 0$ at $t = 0$, and combining with Eq. (2) as follows:

$$A_{Pb210} = \quad \cdot \quad {}_{Pb210} \cdot n_{Pb210} \tag{3}$$

$$= \quad \cdot A_{Rn} \left[1 - \exp\left(- \quad {}_{Pb210} \cdot T \right) \right] \tag{4}$$

$$= \quad \frac{P}{P_a} k_{i\,Rn} \cdot C_{Rn} \left[1 - \exp\left(- \quad {}_{Pb210} \cdot T \right) \right] \tag{5}$$

Eq. (4) or (5) with process pressure P, operating period T, and operation rate τ enables A_{Pb210} to be estimated. Eq (5) requires $k_{i\,Rn}$ and C_{Rn} for such estimation. On the other hand, Eq. (4) requires A_{Rn}. Therefore, two evaluation methods have been developed in this study: the 'principle method' based on the radon adsorption equilibrium coefficient of the adsorbent, and the 'practical method' based on the radon adsorption amount directly determined. The examinations and results for these methods are described.

3. THE PRINCIPLE METHOD

This method evaluates A_{Pb210} based on $k_{i\,Rn}$ calculated by a breakthrough curve obtained by an adsorbent column test using the surrogate gas including a determined concentration of radon.

3.1. Examination to obtain the radon adsorption equilibrium coefficient

Flow diagrams of the test devices are shown in Fig. 1. The upper diagram shows the apparatus for preparation of the surrogate gas, while the lower diagram shows the adsorption test apparatus.

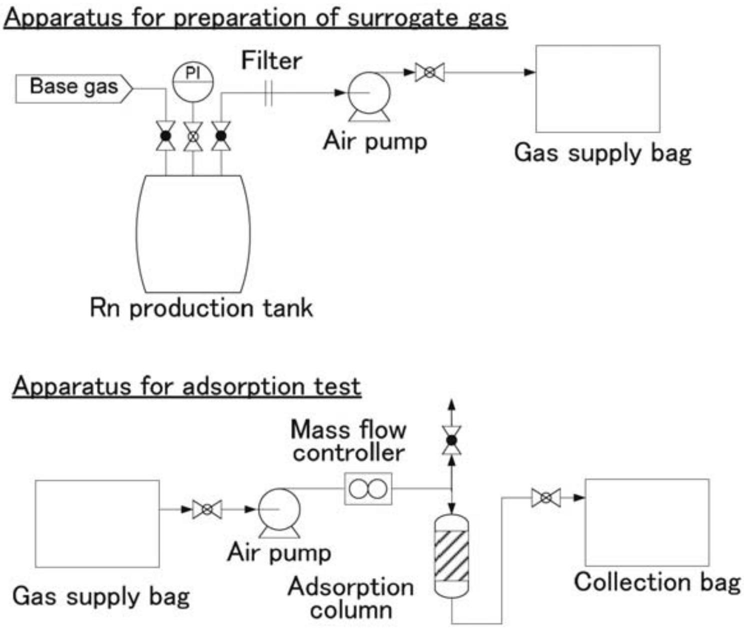

FIG. 1. Test apparatus for obtaining radon adsorption equilibrium coefficient.

The base gas was charged into the radon production tank containing a ^{226}Ra source. When the radon concentration in the base gas reached a certain value, the gas was transferred into a gas supply bag as surrogate gas. The surrogate gas was fed into a column which was packed with adsorbent. The effluent gas from the column was collected in collection bags. The radon concentration of the gas collected in the collection bags and the surrogate gas in the supply bag was measured with a scintillation radon monitor (Pylon Model AB-5).

The breakthrough curve was described as the radon concentration in the effluent gas versus time. The amount of radon adsorbed in the column was obtained as an integral of the difference between the radon concentration in the surrogate gas and that in the effluent gas. The radon adsorption equilibrium coefficient was obtained as a ratio of the radon concentration in the adsorbent to the radon concentration in the surrogate gas. The parameters of the experiments for obtaining the radon adsorption coefficients were as follows:

(a) Adsorbent:
  — Molecular sieve 4A (i = 1),
  — Silica gel (A) (i = 2),
  — Silica gel (B) (i = 3),
  — Mass: 10 g;
(b) Adsorption parameters:
  — Radon concentration $A_{Rn}(e)$: 5000–20 000 Bq/m^3,
  — Adsorption pressure $P(e)$: 0.1 MPa,
  — Adsorption temperature: 293, 303 and 318 K,
  — Flow rate: 63 and 126 mL/min.

Molecular sieve 4A and silica gel (A) are used in actual NG and LNG plants. Silica gel (B) was studied in this investigation but is not used in actual NG and LNG plants. Argon was used as the base gas in this study for safety reasons; argon is also a noble gas that does not interact with radon. The adsorption behaviour of Ar and CH_4 is expected to be similar because the radii of Ar and CH_4 are similar and both Ar and CH_4 are non-polar. Silica gel and a molecular sieve were studied as the adsorbent since silica gel is used as a guard bed and the molecular sieve is used as a dehydration adsorbent in LNG plants.

3.2. Test results

The breakthrough curves obtained are shown in Fig. 2. The result showed that adsorption reached equilibrium in about 60 min. The amount of radon adsorbed was obtained from the breakthrough curves. The relationship between the radon concentration in the feed gas and the amount of radon adsorbed is shown in Fig. 3.

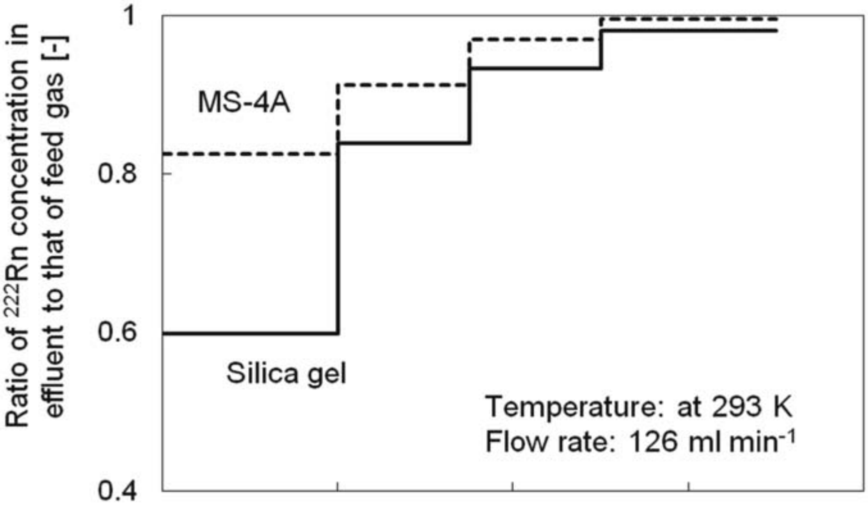

FIG. 2. Radon breakthrough curves of molecular sieve 4A and silica gel (A).

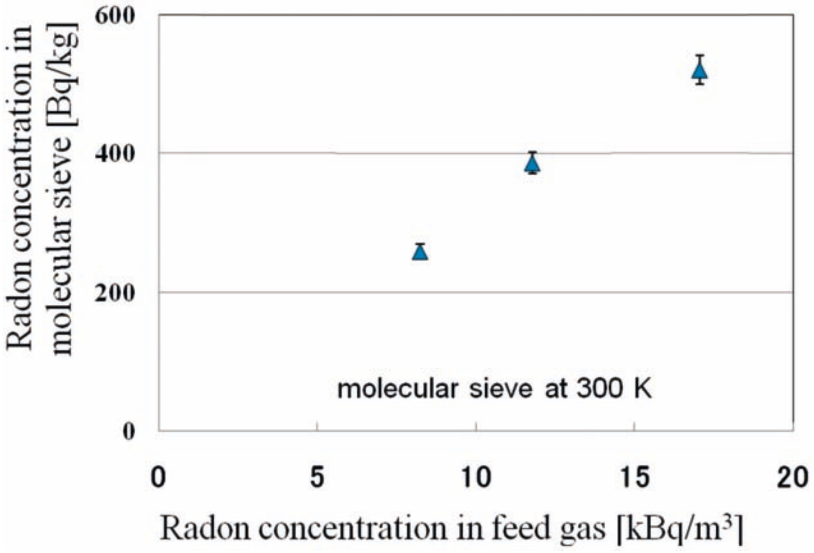

FIG.3. Relationship between radon concentrations in feed gas and in molecular sieve 4A.

It was confirmed that assumption (a) in Section 2 is correct: the amount of radon adsorbed is proportional to the radon concentration in the gas, which means that radon is adsorbed on the molecular sieve and silica gel by physical adsorption. The radon adsorption equilibrium obtained is shown in Fig. 4.

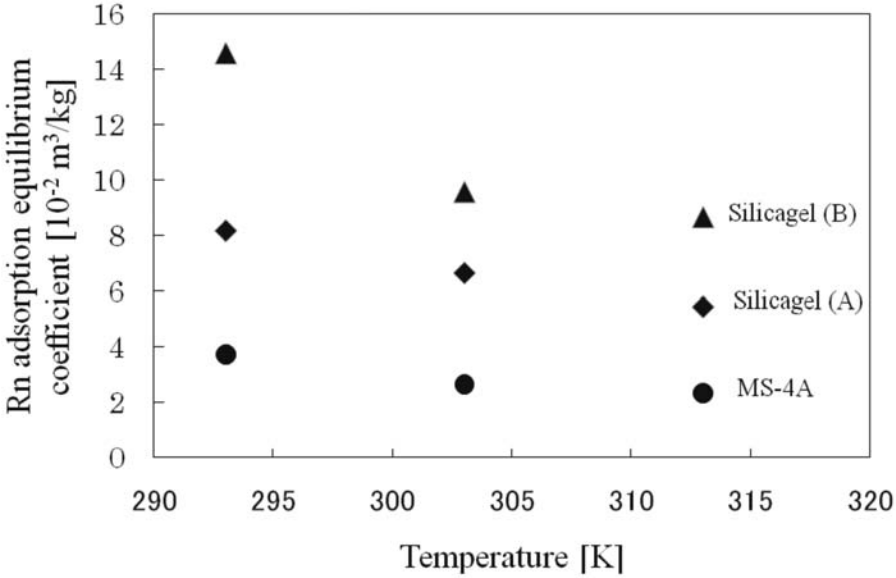

FIG. 4. Radon adsorption equilibrium in silica gel and molecular sieve.

3.3. Estimation of ^{210}Pb accumulation

The accumulation of ^{210}Pb on the adsorbents was estimated, based on the radon adsorption equilibrium at 293 K in Fig. 4. The relationship between the operating period T and the ^{210}Pb concentration in the adsorbent A_{Pb210} was obtained from Eq. (5), where the radon concentration in NG, C_{Rn}, was set at 1000 Bq/m^3 and the operating conditions were P = 5.5 MPa and $\tau = 2/3$. The relationships between T and A_{Pb210} in the molecular sieve 4A ($k_{1\,Rn} = 0.037$) and in the silica gel (A) ($k_{2\,Rn} = 0.076$) are described in Fig. 5, which shows that the value of A_{Pb210} of molecular sieve 4A treating NG with 1000 Bq/m^3 of radon will not reach the exemption level (1 Bq/g) during 30 years of operation. In the case of silica gel, the exemption level will be reached after about 15 years.

The concentration of ^{210}Pb in the adsorbent depends upon the radon concentration in the NG and upon the operating conditions. The relationship between the operating period T (in years) and C_{Rn} was obtained from Eq. (4). The calculation result is shown in Fig. 6. The time period Tc during which A_{Pb210} approaches, but does not exceed, 1 Bq/g (that is, during which the adsorbent can be treated as non-radioactive material) can be deduced from Fig. 6.

It was confirmed that the principle method can predict A_{Pb210} and Tc. However, this method requires a test gas containing the determined concentration of radon, the preparation of which requires special equipment, such as the radon

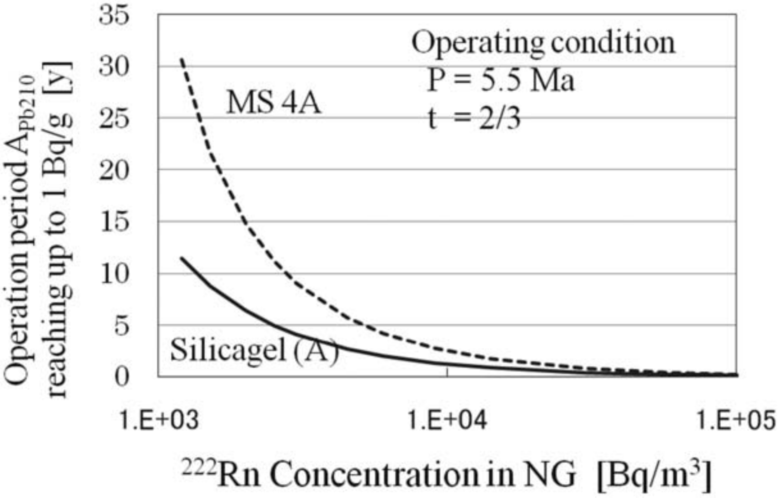

FIG. 5. Relationship between operating period and ^{210}Pb concentration in adsorbents.

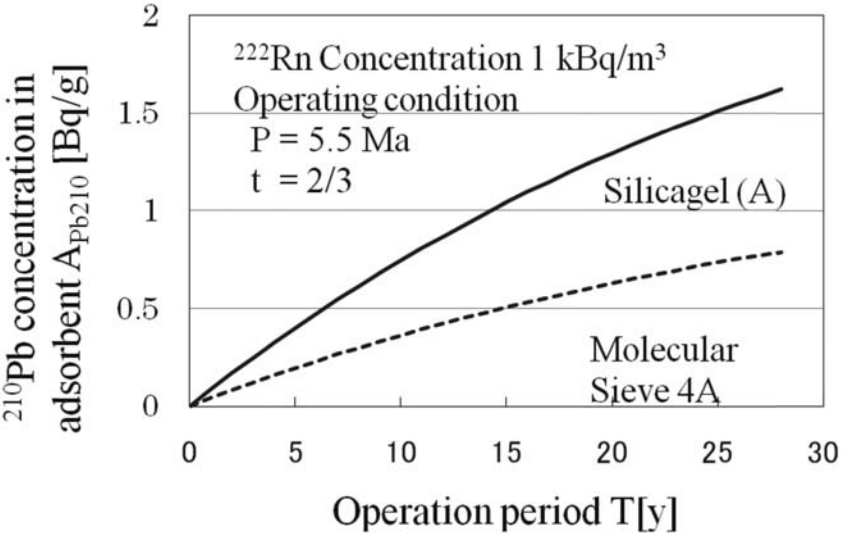

FIG. 6. Relationship between radon concentration in gas and in adsorbents.

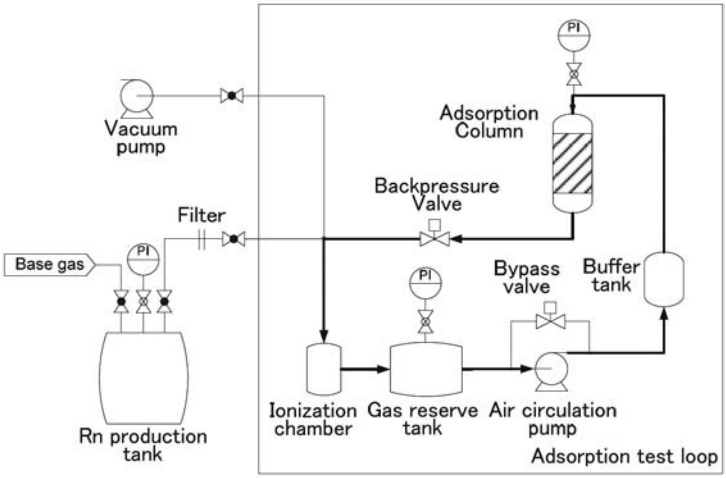

FIG.7. Test apparatus for verifying the method for determining radon adsorption.

monitor and the ^{226}Ra source. Therefore, an easier method not needing special equipment was developed.

4. THE PRACTICAL METHOD

This method evaluates A_{Pb210} based on A_{Rn}, which was determined by an adsorption column test using real NG. The radon adsorption coefficient A_{Rn} is determined by gamma spectrometry for ^{214}Pb and/or ^{214}Bi in the column. The study was carried out to examine the theoretical applicability of the practical method by comparing $k_{3\,Rn}$ obtained by the practical method with that obtained by the principle method.

4.1. Verification of the practical method

A test apparatus for verifying the method of determining the amount of radon adsorption is shown in Fig. 7. The test apparatus consisted of the radon production tank and the adsorption test loop. The adsorption test loop consisted mainly of an adsorption column, a gas reserve tank, a circulation pump and an ionization chamber.

Base gas was charged into the radon production tank containing a ^{226}Ra source. When the radon concentration in the base gas reached a certain value, the gas was transferred into a gas reserve tank as test gas. The test gas was circulated by a pump in the adsorption test loop to supply it to the adsorption column. The adsorption pressure $P(e)$ was controlled by a backpressure valve and a bypass valve. After radon adsorption, equilibrium was established and the adsorption column was isolated and detached from the adsorption test loop while maintaining the adsorption pressure. The concentration of radon in the test gas was determined by an ionization chamber method at atmospheric pressure. The adsorption column was measured by low background gamma spectrometry to determine the ^{214}Pb and/or ^{214}Bi activity. This enabled the radon activity to be determined because ^{214}Pb and ^{214}Bi in the column were in radioactive equilibrium with the radon adsorbed. The parameters of the experiments for obtaining A_{Rn} were as follows:

(a) Adsorbent:
 — Silica gel (B) (I = 3)
 — $k_{3\ Rn}$: 0.15 m^3/kg
 — Mass: 22.4 g

(b) Adsorption parameters:
 — Radon concentration $C_{Rn}(e)$: 8000 Bq/m^3;
 — Adsorption pressure $P(e)$: 0.15 MPa;
 — Adsorption temperature: 293 K.

4.2. Test results

The concentration of ^{214}Bi in the adsorption column was determined to be 1.9 Bq/g, equal to A_{Rn}. The value of $k_{3\ Rn}$ was obtained by dividing A_{Rn} by $[C_{Rn}(e)\cdot P(e)/P]$ and was determined to be 0.24 m^3/kg. This value matched the value of $k_{3\ Rn}$ (0.15 m^3/kg) obtained using the principle method within a factor of 1.6. The result showed that the practical method predicts A_{Pb210} using actual NG without the need for determining radon concentration.

5. CONCLUSION

The following results were confirmed in the laboratory study:

(a) The principle method and the practical method are both capable of estimating the value of A_{Pb210} of the spent adsorbent at the time of change of adsorbent in NG and LNG plants before recovery of the spent adsorbent;

(b) The principle method and the practical method also predict the change interval within which the A_{Pb210} of the spent adsorbent is below the exemption level.

Evaluation methods for ^{210}Pb accumulation on adsorbents in NG and LNG plants have thus been established.

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PRACTICAL ISSUES FOR DEALING WITH NORM IN THE NETHERLANDS

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Abstract

In the Netherlands, NORM is used and produced by different industries. It is partly in the form of raw materials, partly in the form of waste and partly in a form suitable for recycling. The material can be feedstock for processing into products. Some of the materials end up as residues. For residues that cannot be used or recycled there are two options: treatment and storage as radioactive waste or disposal as hazardous waste at a dedicated repository. Treatment of (radioactive) waste and recycling and use of residues are strictly regulated. In this paper an update of the current treatment technologies for NORM by-products and waste is given. Special attention is given to the legislation in the Netherlands as well as practical aspects that cause problems in day-to-day operations. These include measurement problems with parameters such as surface contamination, dust concentration and activity concentration; problems with supervision by a radiation protection expert in small NORM industries; and problems with the treatment of small amounts of NORM waste. The practicability of the Dutch legislation is reviewed (particularly in the light of the forthcoming revision of European Council Directive 96/29/Euratom) and potential solutions and approaches are suggested.

1. INTRODUCTION

As in other countries, a wide variety of NORM is generated by industries in the Netherlands. Some of the material is suitable for use or recycling, while some is considered waste and has to be disposed of. According to the Dutch waste management hierarchy (the so-called ‘ladder of Lansink’) the use and recycling options are the first to be considered. Only if these options are not feasible can the material be disposed of and is (by definition) considered to be waste. In this case, two options are available: disposal at the Central Organization for Radioactive Waste (COVRA) or at an authorized waste repository (landfill) for hazardous waste.

In this paper, a short overview of the current situation concerning types and amounts of NORM in the Netherlands as well as available routes for the recovery or treatment of this material are presented. Relevant legislation and the related opportunities and practical difficulties for the NORM industry are discussed. Specific attention is given to new treatment options developed by COVRA for the conditioning of the material prior to long term storage. Last (but not least), alternative routes that are available or under development for the recycling of NORM are dealt with.

2. LEGISLATION

Separate legislation for NORM was introduced in 2004 with the *Ordinance concerning Natural Activity of Sources of Ionising Radiation* (NABIS) [1]. The revised version of the European Council Directive 96/29/Euratom (the Euratom Directive) [2], currently under development, will change NABIS considerably. The method for determining whether a notification or an authorization is required will change from a one stage approach based on an exemption or clearance criterion, to a two stage approach based on, firstly, an exemption or clearance criterion and, secondly, a dose criterion.

Despite the forthcoming revision of the Euratom Directive, NABIS had already been revised in 2008. The purpose of this revision was to address certain consequential issues and issues of practicality rather than to change the methodology itself, with one exception. For NORM with an activity concentration less than 10 times the exemption level in a quantity of <1 t, it was no longer necessary to have the supervision of a radiation protection expert or to notify the relevant authority when the supplier of the material had already done so.

In the Euratom Directive, the threshold for regulation is 10 kg or less in the case of an activity concentration not exceeding 1 Bq/g for ^{238}U or ^{232}Th series radionuclides in equilibrium. As a result, many small users (for instance, those using zircon sand for pottery) fall under the regulatory regime. Although there is no real risk at these levels of activity and amounts of materials, a large amount of specialist work is needed to comply with legislation. By raising the exemption level to 1 t of NORM, the ‘household’ uses of NORM, such as granite worktops, refractories in open fireplaces, zircon sand for home pottery making, are exempted from legislation. This aspect is even more interesting, as it could (we think it should) be considered in the revision of the Euratom Directive.

3. DESCRIPTION OF SCENARIOS

3.1. Use of NORM residues

Since the option of ‘use’ is the primary target, it is explicitly mentioned in NABIS. In many cases, following this approach involves dilution to make the NORM residue suitable for further use, by mixing it with other components such as binders and fillers. For application in civil engineering, a specific requirement is that the NORM residue is diluted to a level such that it is no longer considered radioactive. In this case dilution is not only a treatment option but also a legislative obligation! The general approach for this route is first to see whether the NORM residue can be processed into a useful product (considering technical, economical and environmental aspects) and, second, to conduct an overall safety assessment of the radiological implications for a given process and application to see whether any significant consequences for health and the environment could occur. Significant consequences may occur during the processing of the residue and during the life cycle of the product. In the latter case the leachability of radionuclides and subsequent doses and contamination levels are aspects which have to be taken into consideration. When all of these aspects are shown to meet the corresponding criteria, the technical development and processing of the NORM residue can proceed. As the recycling of NORM residues is a relatively new activity, experience has been gathered only in recent years. After dilution of the NORM residue, the resulting products are no longer considered to be radioactive and are therefore no longer subject to radioactive waste legislation. As a consequence, no real implementation difficulties have occurred. It is expected, however, that questions about the use of NORM residues in this manner, concerning the complete life cycle (including future use), will be raised by both producers and users. In this regard, it is expected that initial studies will be performed within the next few years concerning both the application aspects (construction and maintenance phase) as well as the demolition phase.

As mentioned in previous papers [3, 4], several applications involving the use of NORM residues have been developed in the Netherlands. Although, as stated above, the use and recycling options are emphasized and promoted in NABIS, there are practical limitations to these options. For example only small supplies of NORM can follow these routes.

3.2. Treatment of small amounts of NORM

There is a well developed structure for treatment routes for all types of NORM residues in the Netherlands. However, the choice between routes is determined not only by legislation and infrastructure but also by economics. Because of the economics of recovery, a supply of NORM on a regular basis is needed in order to use this route in practice and, for small amounts of NORM, it may be very difficult to find a solution.

For example, in some cases only minor amounts of zircon sand (typically about 25 t) with elevated levels of radionuclides of natural origin have been found in the Netherlands. Bringing the material to COVRA involves substantial costs. Attempts to find users of zircon sand were unsuccessful since the users of such raw material require high quality material. Moreover, they could not justify the necessary amount of work and (internal) discussion for just a one time acceptance of radioactive material with questionable economic return.

Dilution with other non-radioactive minerals in order to use NORM residues in concrete, for instance, becomes problematic for the same reasons, because no mixing plant operators wish to contaminate their installations with NORM, just for a one time gain. A practical solution for small quantities is very difficult, not because there are no technical solutions, nor due to the fact that legislation forbids these options, but simply because of the economics (and also the perceptions of industries not normally associated with radioactivity). A solution for these types of materials could be a dedicated mixing plant in which all kinds of NORM residue can be mixed with other constituents. One such example is the production of low quality concrete which could be used to make a basic product such as separation blocks used in the storage of bulk granular materials. Recently a private enterprise has applied for a licence for such a mixing operation.

3.3. Waste treatment

If the use of a NORM residue is not possible or feasible, only the option of disposal remains. As pointed out in a previous paper [5], Dutch legislation stipulates that a licensee can only dispose of its radioactive waste (including NORM waste) by handing it over to an authorized organization. For waste in which the activity concentration is more than 10 times the exemption level, only COVRA is authorized for such disposal. Although COVRA is also authorized for accepting low level waste, there is an alternative in the form of specific hazardous waste repositories.

All activities with radioactive substances, excluding shipments, have to comply with the rules laid down in the Radiation Protection Decree, which is part of the Nuclear Energy Act. As far as COVRA is concerned, the following aspects have to be addressed:

(a) Stable material: Because the waste supply is heterogeneous (from over 200 different sources), it has to be treated in order to produce a single final material which complies with the future requirements of the repository. This implies that a wide range of waste types must be processed into a single waste (or a few at most) compatible with the specifications of the envisaged repository and that are easy to handle and maintain.

(b) Acceptable cost: The waste fees must cover all the costs involved, namely treatment, storage and final disposal.

(c) Industrial safety: Radioactive waste treatment and storage are considered normal industrial activities. As a consequence, COVRA is located in an industrial area in the south-west corner of the Netherlands, close to the Borssele Nuclear Power Plant (see Fig. 1). This puts additional demands on safety compared with dedicated sites in more remote locations.

FIG. 1. The COVRA site. (1) Office building and exhibition centre; (2) Building for the treatment of low and medium level waste (AVG); (3) Storage building for conditioned low and medium level waste (LOG); (4) Storage building for high level waste (HABOG); (5) Storage building for low level material from the enrichment industry (VOG); (6) Storage building for low level NORM waste from the ore processing industry (COG); (7) Storage building for scrap material (SOG).

The waste is stored in discrete packages that are resistant to degradation and hazards and can be inspected and retrieved for final disposal. Application of passive safety principles also leads to more predictable costs — the need for control and safety systems, maintenance, monitoring and human intervention is minimized and the costs are largely determined by the capital investments such as equipment and buildings.

As explained in previous papers [5–7], COVRA operates several treatment facilities for the processing of all types of waste into the required solidified form. From the experience gained over the last few years of processing these wastes (ranging from filter cloths and scales to sludges and calcinate), it can be concluded that the available treatment technologies are capable of (pre)treating each of these waste streams. Since, for low level NORM wastes, an alternative (and cheaper) option exists, only small amounts of these types of waste were brought to COVRA, except as described in Section 3.4.

3.4. Disposal at a hazardous waste repository

Before the implementation of NABIS, some NORM wastes were transferred to COVRA. Now they are disposed of at a (licensed) conventional repository for hazardous waste (see Fig. 2). The calcinate produced during thermal phosphorus production is an example of a shift in NORM waste streams to simpler and (therefore cheaper) options. However, according to the environmental licence of such a repository, the chemical composition of the waste streams has to be determined (and also the leachability in the case of high

FIG. 2. Disposal facility for bulk waste at the A & G company.

concentrations of contaminants such as heavy metals). For small amounts of NORM waste, these measurements are relatively costly. Furthermore, where NORM is strongly embedded in the very structure of a waste form, such as in the case of minerals used as fire protection material in old safes, it is almost impossible to extract the samples needed to perform the required measurements. Therefore the authorities accept that for small waste streams (for instance, up to 15 t/a) no measurements of the concentrations of heavy metals and their leachability need be carried out. The argument is that for those small amounts of waste, the total contribution of heavy metals is small compared to the large waste streams accepted by the repository — these measurement requirements were originally intended for waste in quantities of thousands of tonnes per year.

4. REGULATORY AND PRACTICAL ASPECTS

An additional practical aspect of NORM residue treatment is the need for supervision by a radiation protection expert (RPE) who, in the Dutch system, is classified as a ‘level 3 expert in radiation protection’. On the one hand, NORM is generally an open source with the potential risk of the spread of contamination. In NABIS, therefore, the RPE has well defined tasks such as the assessment of risks, the establishment of radiation protection procedures and measures and the provision of information to workers on how to protect themselves. On the other hand, NORM is typically present only in small amounts and/or at small companies. Since in neither case does it make sense to have a full time RPE on site, NABIS allows external RPEs to do the radiation protection work as required by law. A radiation protection monitor (RPM), a ‘level 5B expert’, is needed for performing radiological measurements at the facility unless sufficient surveillance of the RPE and written procedures are in place. However, although this is a practical solution, it sometimes gives rise to discussions between the authorities and the companies that use an external RPE. The main discussion is not so much about assessments or choices for the means of protection, but about the controlling (and informing) function of the RPE (for instance, controlling the register in which the activity levels of the incoming and outgoing NORM are recorded).

From experience, the authors are convinced that by proper instruction of the relevant staff and the involvement of the resident health, safety and environmental (HSE) manager in this matter, normally a good level of radiation protection can be achieved. Moreover, in many cases the HSE measures are exactly the same as those resulting from the assessment of the RPE. With proper support and instruction of an external RPE, an HSE manager is capable of controlling the correct application of protective measures.

5. ACTIVITY CONCENTRATION AND SURFACE CONTAMINATION MEASUREMENTS ON NORM

In NABIS, methods for the calculation of the weighted sum of NORM activity and for the assessment of the activity concentration are prescribed. Annex 4 of NABIS contains a prescribed method for measuring surface contamination. NORM will often contain radionuclides from the same decay chain and NABIS distinguishes between three situations:

(a) The progeny radionuclides are in equilibrium with the parent;
(b) The progeny radionuclides are enriched with respect to the parent;
(c) The progeny are depleted with respect to the parent.

Each situation has been assessed in detail and offers the NORM industry some flexibility in analysing samples and reporting the results, thus allowing it to select the most cost-effective method. It is evident that the assessment of activity concentrations has to be based on gamma emissions and so NORM samples have to be measured using a gamma spectroscopy system. Such a gamma spectroscopy system has to be specially designed for NORM [8] and the recorded spectra have to be analysed in a manner that takes account of parent/progeny ratios and self absorption. Surface contamination measurement methods for NORM are more strictly prescribed. The exemption and clearance criteria applied to NORM are:

(1) 4 Bq/cm^2 averaged over a surface area of 300 cm^2;
(2) For semi-cylindrical objects less than 150 mm in diameter, 4 Bq/cm^2 averaged over a surface area of 1000 cm^2.

Before and during the measurements, the RPM has to ensure that the measurements are not disturbed by variations in the background radiation level and by local magnetic fields. The technical specifications of the measuring equipment have to be such as to ensure a sensitivity for $E_{\beta max}$ of 150 keV or higher, a detection sensitivity better than 0.5 Bq/cm^2 for a single measurement and a measurement deviation of less than 10% or not larger than 1 Bq/cm^2.

6. CONCLUSIONS

The Dutch legislation NABIS has made provision, since 2004, for NORM residues to be used or recycled as useful products or diluted to below the exemption level. NABIS includes methods for the calculation of the weighted sum of NORM activity, for the assessment of the activity concentration and for the measurement of surface contamination.

For NORM industries, an external RPE is required by NABIS and an RPM for performing daily radiological measurements. With proper instruction of relevant staff by the RPE, and involving the resident HSE manager, a good level of radiation protection can normally be achieved.

COVRA is responsible for managing NORM waste designated in Dutch law as radioactive waste (that is, waste for which the exemption level is exceeded by more than 10 times). COVRA is licensed to operate a treatment and storage facility for radioactive waste, including NORM waste.

The disposal, at hazardous waste repositories, of hazardous NORM waste (that is, waste for which the exemption level is exceeded by less than 10 times) is a well known practice in the Netherlands and there are several landfills available. For small users (up to 15 t of NORM waste per year) no measurements of the concentrations of heavy metals and their leachability are required. As a result of the increase in the exemption level for NORM to 1 t in 2008, the ‘household’ uses of NORM are now exempted from legislation.

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ENVIRONMENTAL BURDEN OF NORM RESIDUES

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Abstract

The monitoring and prevention of occupational radiation risk caused by NORM has in many cases become obligatory for industries of concern. Usually, the degree of control applied in such cases has been based on conventional radiation protection standards as applied to radiation risks from artificial sources. Case specific risk scenarios have been developed and have become readily available to industry operators. Far less attention has been paid to the environmental burden of NORM residues that have been designated as waste. Such waste is often deposited directly into the environment, a practice that is forbidden in the case of radioactive waste containing radionuclides of artificial origin. Hence, to assess the environmental impact, it is essential to have information on the radionuclide species deposited, interactions within affected ecosystems and the time dependent distribution of radionuclide species influencing mobility and biological uptake. Preliminary data have indicated a strong need to consider such waste as a specific type of environmental hazard. But NORM residues differ significantly in quantity and quality from ‘classical’ nuclear materials and, moreover, the derived radiation risk is usually associated with the risk caused by other pollutants. That is why they cannot be controlled by applying directly the standards designed and developed for radioactive waste containing radionuclides of artificial origin. Such a situation results in there being no precise regulations in this area and the non-nuclear industry is not aware of the environmental problems caused by NORM residues. This paper discusses all aspects of the radiation risk to the environment caused by NORM residue deposits that have not been regulated from a radiation protection point of view. The inconclusive nature of existing recommendations and models of radiation risk assessment are underlined. General terms such as the environmental effect, and the basic parameters necessary to carry out a consistent and comparable environmental risk assessment have been developed and defined.

1. INTRODUCTION

Radioactivity is a primordial feature of matter and the human environment. Since life emerged on Earth, every living organism has been exposed to ionizing radiation. According to current radiation protection standards, radiation emitted by primordial radionuclides in their natural state that has not been altered due to human activity is not considered to be a source of harmful effects, whether for humans or the environment. There are many areas in the world where the so called ‘natural background’ is elevated as a result of the geological or geochemical structure of the rocks. Whether or not such background radiation can cause a negative or positive effect on humans is a matter of opinion, but if the concentrations of radionuclides of natural origin have been changed as a result of the deliberate or accidental actions of humans, it is quite another matter. Such alteration of the natural state can result in an incremental radiation risk to humans as well as to the environment and non-human biota. Hence, the monitoring and prevention of the occupational radiation risk caused by NORM has in many cases become obligatory for industries of concern. Unfortunately, much less attention has been given to the environmental burden of residues deposited as waste and containing enhanced concentrations of radionuclides of natural origin. There is a wealth of data relating to the behaviour of radionuclides of natural origin in their natural state on the borderline between abiotic and biotic matter. Frequently, the processes of metabolism lead to concentration of some long lived radionuclides of natural origin in particular tissues of fungi, plants as well as animals [1]. The derived committed dose can be higher than doses resulting from radionuclides of artificial origin accumulated simultaneously [2]. There are some examples of societies based on a limited trophic chain where the related committed dose to individuals caused by radionuclides such as polonium and lead being biologically accumulated by ingestion is significant. Annual effective doses from ^{210}Po as high as 10 mSv have been reported for some northern aboriginal communities consuming large amounts of caribou [3]. Annual doses reaching 3 mSv were reported for a population of fishermen living only on seafood [4, 5]. If such processes can happen in an unmodified environment one can easily imagine what might happen in the vicinity of a NORM residue deposit. Despite that, the environmental aspects of NORM residues are never discussed and reported. In the light of the extremely conservative approach to the management of ‘classical’ radioactive waste, such as nuclear waste, such a situation regarding NORM waste seems strange at the least.

The activity concentration of radionuclides of natural origin in NORM type waste (NTW) is sometimes high enough to classify the material as radioactive waste. In spite of that, such material is often deposited directly into the environment, something that is strictly forbidden in the case of radioactive waste. For this reason, the assessment of the environmental impact of contamination by radionuclides of natural origin requires information on the radionuclide species deposited, interactions within affected ecosystems and the time dependent distribution of radionuclide species influencing the mobility and biological uptake [6]. Also, the definition of the expected effect on the environment and how it should be evaluated is crucial for the planning of suitable regulation.

2. NORM TYPE WASTE CHARACTERIZATION

Usually, the total amount of NTW collected in one deposit reaches hundreds of thousands of cubic metres or tonnes. Quite often, either the radionuclide activity concentration or the total activity is high enough for such waste to be classified as radioactive waste. It has been common practice for such kinds of waste to be deposited in heaps or piles, without any means of protection and also without regard to radiation protection requirements, if applicable. The exposure to meteorological processes such as a rainfall or wind can cause uncontrolled migration of contaminated waste into every compartment of the environment. Also, NTW placed directly into the environment can initiate some additional chemical or physical processes, leading to the selective transfer and accumulation of particular radionuclides and disequilibrium in the uranium and/or thorium decay series. This can result in severe disturbance to the surrounding environment. From a radiation protection point of view, the main properties of NTW that differentiate it from ‘classical’ radioactive waste are:

(a) Their occurrence in bulk quantities deposited directly in the environment;
(b) The wide variety of chemical compounds and different mineral contents;
(c) The possible co-existence of other pollutants such as heavy metals, sulphates and hydrocarbons.

NTW produced by non-nuclear industrial activities such as mining, mineral processing and coal fired power generation contains several long lived radionuclides of natural origin from the uranium and thorium decay series. Taking such radionuclides into consideration and either their activity concentration or total activity, some NTW should be classified as ‘radioactive waste containing alpha emitters’. An activity concentration criterion sometimes used for such a classification is 10 Bq/g for radium isotopes, for example, but in practice the decision to classify NTW as radioactive waste, alpha emitting or not, is rarely taken. This is a consequence of the technical problems and financial implications that would follow such a decision, bearing in mind that such waste must be sealed and disposed of in a repository which, in the case of alpha emitters, should be a deep underground repository. Classification of NTW as radioactive waste would have enormous cost implications and would result in available repositories becoming filled very quickly. There is no possibility of treating such waste in terms of existing regulations, which have been established for dealing with nuclear waste. Furthermore, there is no special regulatory provision and no accepted manner of disposal. From an environmental point of view, it is interesting to consider the situation of waste with a radioactivity content slightly lower than the criterion for classifying it as radioactive waste, for example 9 Bq/g in the case of a 10 Bq/g criterion for radium isotopes. The environmental impact of waste with a radium activity concentration of 9 Bq/g is unlikely to be significantly different from that when the activity concentration is 10 Bq/g. The clearance criterion recommended in international standards for radionuclides in the uranium and thorium decay series is 1 Bq/g [7]. Consequently, even following existing regulatory guidance, a big ‘gap’ between 1 and 10 Bq/g quickly becomes evident.

After deposition of NTW in the environment, the elevated concentrations of radionuclides of natural origin will, firstly, result in enhanced exposure of biota to external gamma radiation. Therefore, an accurate knowledge of the total radionuclide concentration in such material is needed in order to assess the potential radiological risk at the site concerned. However, knowledge of the physical presence of every radionuclide would provide only a part of the knowledge necessary to evaluate its potential for harm. The radiological hazards can be increased by the migration of the mobile fraction of these radionuclides into the surrounding environment [8]. After release into an ecosystem, such radionuclides can migrate and then interact with the processes occurring on the borderline of the abiotic environment and biota. For this reason, it is necessary to know the mobility of radionuclides [9]. The mobility and environmental behaviour of every element depend on its speciation in certain waste material [10]. The speciation of a radionuclide is generally related to its physical and chemical form, that is, to the existence of simple and complex ions in interstitial solution and exchangeable ions associated with waste material organic fractions occluded or co-precipitated with metal oxides, carbonates, sulphates and other secondary minerals.

The exposure to external gamma radiation is only the tip of the iceberg. One should remember that in the ^{238}U and ^{232}Th decay series there are 8 and 6 alpha emitters, respectively (with an additional 7 in the ^{235}U series). Even considering the decay chain segments starting from radium (radium ^{226}Ra and ^{228}Ra, respectively), a common situation with NTW, the number of alpha emitters in each chain segment decreases only to 5. In the case of plants, especially plant roots, the exposure to external alpa radiation is as important as exposure to alpha radiation emitted by incorporated emitters, but this is hardly ever taken into consideration. Also, beta emitters should be taken into account. The weighting factors for alpha and beta radiation established for humans cannot be directly applied to plants but there is no rational reason why they should be significantly lower. Direct measurements have shown that the absorbed dose resulting from alpha radiation can reach the same level as that from gamma radiation after applying weighting factors [11].

Besides the problems caused by activity concentration, radionuclide speciation and migration, the evolution of the risk scenario must be taken into consideration. The most common radionuclides responsible for risk creation are radium isotopes. When the source of NTW is formation water, such as in the oil and gas industry and underground mining, radium isotopes from both the uranium and thorium series (^{226}Ra and ^{228}Ra) predominate. This implies that there is significant disequilibrium in the natural decay series. Immediately after deposition of the material, the contaminant (besides possible other, non-radioactive contaminants) is almost entirely radium. Radium, being usually only weakly mobile, has a low bio-availability in the environment. It has been demonstrated that radium either does not create soluble radium–barium sulphate or its atoms remain strongly bonded within fine clay minerals. The bio-available part usually does not exceed 1% of the total activity [12]. Consequently, the environmental risk is limited only to exposure to external gamma radiation, or, as shown above, to alpha radiation and can be partially limited by the application of a cover made from an inert material. Moreover, since the half-life of ^{228}Ra is 5.7 a, the exposure to this radionuclide will decrease relatively quickly if it is not supported by its parent radionuclide. The only difficulty is that, for a decay chain segment headed by unsupported ^{228}Ra, the activity concentration of progeny such as ^{224}Ra can exceed the activity concentration of the parent after a few years. This can make measurement and dose assessment complicated.

Quite a different situation exists for ^{226}Ra. Its half-life (1600 a) is long, not only from a human perspective but also from the perspective of the environment and changes in the ecosystem. As already mentioned, the bio-availability of radium is very weak, but such a comfortable situation with immobilized radium does not last long. Taking into consideration the characteristics of the uranium decay chain, a slow but nevertheless permanent ingrowth of the long lived progeny ^{210}Pb and ^{210}Po can be expected. These radionuclides are well known for their easy migration in the environment and their availability to biota. Radium isotopes, as they decay, become released from barium sulphate or clay mineral structures. Moreover, the gases radon and thoron are produced as progeny of radium, above lead and polonium in the relevant decay chain. At sites where radium containing NTW has been deposited, one should expect to find, after 100 years, lead and polonium isotopes in a mobile and bio-available form and at the same activity concentrations as their parent radium isotopes. Apart from their specific chemical properties, these lead and polonium isotopes are beta and alpha emitters, respectively, and both are chemically toxic. In summary, NTW, even though appearing to be not particularly hazardous at present, can be regarded as potentially posing a hazard in the future.

3. ENVIRONMENTAL EFFECT

Many scientific publications include the term ‘environmental effect’ or ‘environmental impact’ in their title. While most of them are concerned with radionuclides of artificial origin deposited in the environment mainly as a result of an accident or from the testing or use of nuclear weapons, the important point is that, up to now, the term environmental effect has not been defined. Such publications hardly ever go beyond the measurement of activity concentration in a particular compartment of the environment. Some deal with soil-to-biota transfer factors, but are usually limited to contamination of the human trophic chain and assessment of committed doses without considering environmental effects. Recent recommendations of the International Commission on Radiological Protection [13] introduce a reductionistic approach based on early mortality, morbidity and reduced reproductive success of ‘reference organisms’. Such a concept, focused on individuals, is contrary to the general principle of radiation protection concerning non-human biota. In the case of the environment, there is no obligation to protect individuals or even species of non-human biota, but rather the environment in general. The environment can mean either the local or the worldwide ecosystem, depending on the scale of the event subject to assessment. The ecosystem can be understood as a set of processes going on in the environment. The basic approach considers only the circulation of matter and energy within the abiotic environment and different trophic levels of living organisms. From this point of view, the mortality, morbidity and reduced reproductive success of individuals playing a role in this process can be treated as environmental effects. The relationship between the absorbed dose and such observed environmental effects could be a good tool for assessing the environmental effect of the pollutant of concern, but such effects are currently limited only to deterministic effects as defined for humans.

However, if one considers the ecosystem thoroughly, especially from the point of view of the flow of time rather than of matter and energy, another set of processes going on in the ecosystem can be noticed, namely, the processing of genetic information as a result of the flow of time. The ecosystem is not stable — the process of evolution is still continuing (see Fig. 1). Changes in the surrounding circumstances create pressure on the processing of genetic information by biota. On the other hand, each change in the code of life influences the adaptation to new conditions. Feedback between biota and the environment exists. All factors influencing the process of passing on information, even very small factors, can give either an advantage or a disadvantage to some

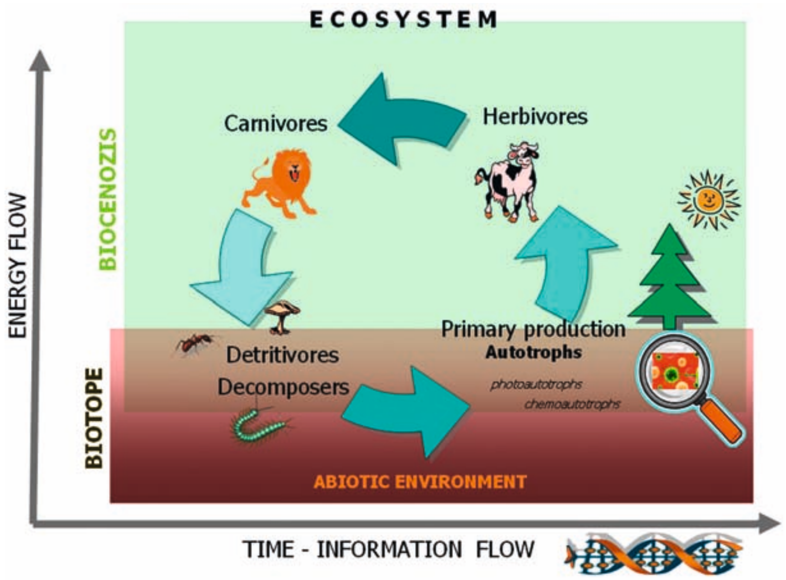

FIG. 1. The processes going on in an ecosystem.

individuals or species and cause them to become dominating (or suppressed) after only a few generations.

For the reasons explained in Section 2, radioactivity can have an effect on the ecosystem. The influence of NTW can be significant and highly probable because of the direct contact with the environment over a period long enough for interactions to occur. The problem is how to measure and quantify the possible effect. It is clearly too late to observe morbidity and especially mortality for this purpose. The interaction between radiation and a living organism starts at the cellular level. Cell damage is the first manifestation of the detrimental effects of radiation. All further effects are merely a continuation of the interaction process. At the molecular level, there are two different effects observed: cytotoxicity and genotoxicity. The former can be expressed as a deterministic effect, while the second can be regarded as a disturbance to the flow of information within biota. The relation between radionuclide activity concentration, absorbed dose and effects at the molecular level seems to be the best tool for the assessment the actual condition of biota exposed to NTW (see Fig. 2). *Quantified effects at the molecular level and the probability of their occurrence is the best measure of the so-called environmental risk.* However, in the case of NTW, associated pollution often exists, making it difficult to identify which kind of pollutant is directly responsible for the observed effect. There is also the possibility of either synergistic or antagonistic effects.

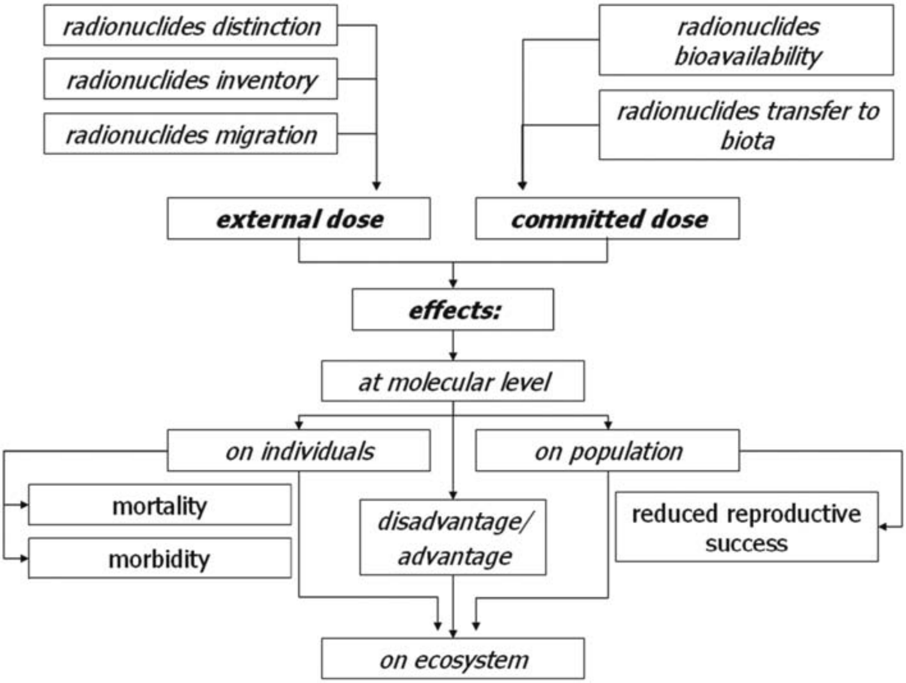

FIG. 2. The relationship between parameters describing environmental effects.

Measurement and assessment of absorbed doses and concentrations of other pollutants in the environment could provide a detailed description of the ecological situation, but they provide only indirect indications of the potential biological consequences. Biological assays, in turn, provide an integrated estimation of the toxicity and genotoxicity of all existing environmental pollutants. A combination of these two techniques enables the identification of the major sources of environmental risk that require continuous biological monitoring. An adequate assessment of the risk to the environment from contamination needs to be based on the simultaneous use of the traditional monitoring of radioactivity levels and genotoxicity tests.

4. CONCLUSIONS

An adequate environment quality assessment cannot rely only on information about pollutant concentrations. It is therefore impossible to estimate the environmental risk due to the action of a combination of agents based only on the knowledge of their concentrations in the environment. This conclusion emphasizes the need to update some of the current principles of environmental standardization which are still in use nowadays (for instance, universal clearance levels). With this in mind, the determination of whether the presence of NTW can cause a detriment to the environment needs to involve a multi-stage evaluation process culminating in the identification of the real effects at an appropriate level. The process should result in the formulation of the relationships between:

(a) Occurrence and exposure;
(b) Exposure and dose;
(c) Dose and effects on non-human species (representative species or the whole ecosystem).

Dedicated investigations should be sequentially focused on all stages of the risk creation scenario.

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A MATHEMATICAL TOOL FOR SIMULATING THE DISPERSION OF NORM RELEASES IN THE MARINE ENVIRONMENT: APPLICATION TO THE WESTERN ALBORAN SEA[1]

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Abstract

A numerical model which simulates the dispersion of radionuclides of natural origin in the marine environment has been developed and applied to the western Alboran Sea. The model consists of a hydrodynamic module which operates off-line, and the dispersion module itself. The hydrodynamic module consists of two models: a 2D barotropic model which provides instantaneous tidal currents and a 3D baroclinic model which provides the long term residual currents. The calculated currents are stored in files which are later read by the dispersion model. The calculated tidal and residual currents have been compared with measurements in the area. In particular, the well known Western Alboran Gyre is reproduced by the model. The dispersion model essentially solves the advection–diffusion equation using finite difference techniques. Interactions of dissolved radionuclides with suspended particles have been neglected in this case, given the low particle concentrations present in the western Alboran Sea and Strait of Gibraltar. The model has been applied to simulate ^{226}Ra releases from hypothetical phosphate fertilizer industries located on the Spanish and Moroccan coasts. It is a useful tool for assessing the effects of planned releases of radionuclides of natural origin in the marine environment.

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1. INTRODUCTION

The area comprising the Strait of Gibraltar and the Alboran Sea constitutes the connection between the Mediterranean Sea and the Atlantic Ocean and has been the subject of many radioecological studies [1]. The water dynamic is characterized by a surface inflow of Atlantic water and a deep outflow of dense Mediterranean water. The Atlantic jet flows along the Spanish coast and curves to the south to incorporate the so-called Western Alboran Gyre (WAG) [2]. The objective of this work was to construct a dispersion model covering the Strait of Gibraltar and the Alboran Sea that was able to simulate ^{226}Ra releases from hypothetical phosphate fertilizer industries located on the Spanish and Moroccan coasts.

2. MODEL DESCRIPTION

The topography of the area covered by the model can be seen in Fig. 1. The computational domain extends from 35º00’N to 36º74’N and from 05º35’W to 03º00’W. Water depths were downloaded from the NOAA Geodas database having a resolution of $\Delta x = 3032$ m and $\Delta y = 3712$ m.

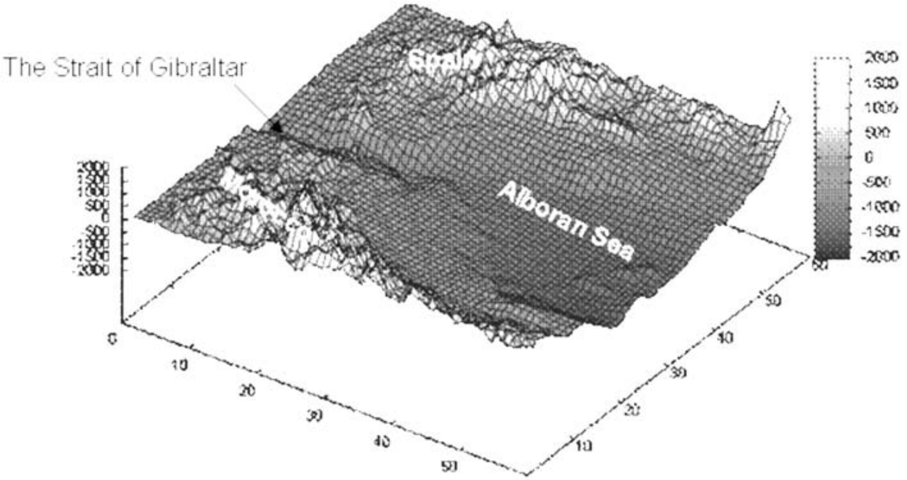

FIG. 1. Topography of the area covered by the model.

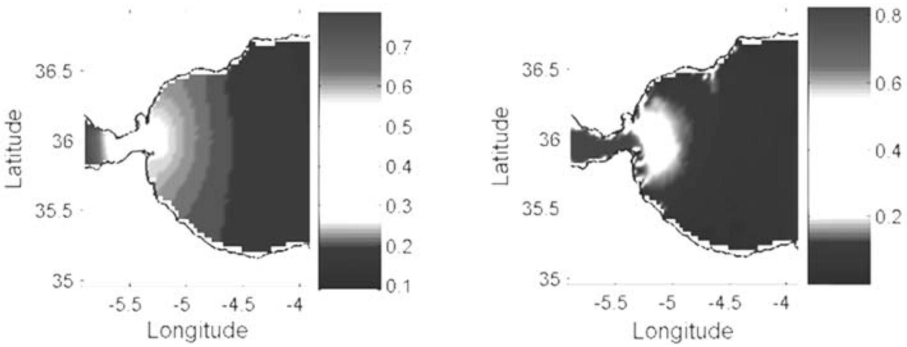

FIG. 2. Tide amplitude (left) and current amplitude (right) for the M_2 tide computed by the 2D depth averaged model [3].

2.1. Hydrodynamics

Tidal currents in the area covered by the model were obtained by means of a 2D depth averaged model [3]. The model solves the standard equations and provides the water currents at each compartment of the calculation mesh and at each time step. Currents are treated by standard tidal analysis to obtain tidal constants that are stored in files that will be read by the dispersion code. Tide and current amplitudes computed by the model are presented in Fig. 2 and are in good agreement with measurements and earlier computations.

The 3D hydrodynamic equations are the following [4]:

$$\frac{\partial}{\partial t}+\frac{\partial}{\partial x}\left(\int u\cdot dz\right)+\frac{\partial}{\partial y}\left(\int v\cdot dz\right) \quad = \quad 0$$

$$\frac{\partial u}{\partial t}+u\cdot\frac{\partial u}{\partial x}+v\cdot\frac{\partial u}{\partial y}-\Omega\cdot v+g\cdot\frac{\partial}{\partial x}+\frac{g}{{}_0}\cdot\int_z\frac{\partial \;{}_w}{\partial x}\cdot dz$$

$$=\frac{\partial}{\partial z}\left[K\cdot\frac{\partial u}{\partial z}\right]+A\cdot\left[\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}\right]$$

$$\frac{\partial v}{\partial t}+u\cdot\frac{\partial v}{\partial x}+v\cdot\frac{\partial v}{\partial y}-\Omega\cdot u+g\cdot\frac{\partial}{\partial y}+\frac{g}{{}_0}\cdot\int_z\frac{\partial \;{}_w}{\partial y}\cdot dz$$

$$=\frac{\partial}{\partial z}\left[K\cdot\frac{\partial v}{\partial z}\right]+A\cdot\left[\frac{\partial^2 v}{\partial x^2}+\frac{\partial^2 v}{\partial y^2}\right] \qquad (1)$$

- where ξ is the water elevation above the mean level, g is the acceleration due to gravity, Ω is the Coriolis parameter, u and v are the two components of the velocity, ρ_w is the water density and K and A are respectively the vertical and horizontal eddy viscosities. The vertical component of the water velocity can be obtained from the continuity equation:

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}+\frac{\partial w}{\partial z}=0 \qquad (2)$$

The water salinity and temperature are also calculated all over the computational domain by solving the advection–diffusion equations:

$$\frac{\partial S}{\partial t}+u\cdot\frac{\partial S}{\partial x}+v\cdot\frac{\partial S}{\partial y}+w\cdot\frac{\partial S}{\partial z}=\frac{\partial}{\partial z}\left[K_v\cdot\frac{\partial S}{\partial z}\right]+K_h\cdot\left[\frac{\partial^2 S}{\partial x^2}+\frac{\partial^2 S}{\partial y^2}\right]$$

$$\frac{\partial T}{\partial t}+u\cdot\frac{\partial T}{\partial x}+v\cdot\frac{\partial T}{\partial y}+w\cdot\frac{\partial T}{\partial z}=\frac{\partial}{\partial z}\left[K_v\cdot\frac{\partial T}{\partial z}\right]+K_h\cdot\left[\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial y^2}\right] \qquad (3)$$

where K_h and K_v are horizontal and vertical diffusion coefficients. The density of the water is determined from the equation of state and the turbulent viscosity is calculated by solving the turbulent kinetic energy equation [5].

All equations were solved using an explicit finite difference scheme by providing the suitable boundary conditions for seawater elevation, currents, salinity and kinetic energy. The components of the wind and bottom stresses and no flux for salinity were specified as boundary conditions at the sea surface bottom. On the other hand, along the open boundaries, a radiation condition is applied for the water velocity component normal to the boundary [6]. All the parameters in the equations have been fixed according to the corresponding values found in the literature and some of them were established by trial and error.

To solve the hydrodynamic equations, a time step of Δt =10 s was adopted according to the stability criterion CFL (Current-Friederichs-Lewy). Discretization in the vertical direction consisted of dividing the water column into 35 layers with a fine resolution of 20 m for the first 30 layers and 300 m for the rest until the bottom. Boundary conditions for water fluxes were 1.25 sverdrup (1 sverdrup = 10 m^3/s) for the incoming flux from the Atlantic Ocean through the Strait of Gibraltar, and 1.20 sverdrup for the outgoing flux near the bottom [7]. The solution of the 3D hydrodynamic equations provides the components of residual water currents, salinity, temperature and viscosity distributions at each

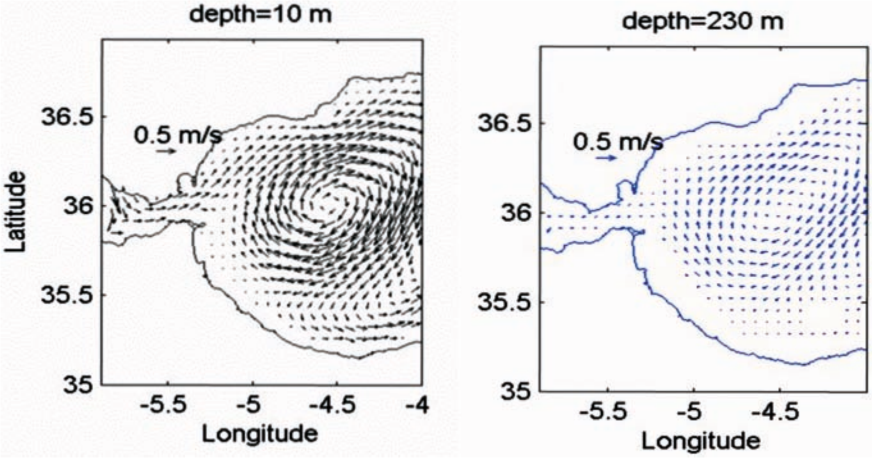

FIG. 3. Residual currents at two different depths.

box within the calculation mesh. Fig. 3 shows the currents computed by the model at two different depths (10 m and 230 m) after a simulation of 50 d.

The hydrodynamic model reproduces in a satisfactory way the overall circulation of water in the application area. The jet of Atlantic water, which enters via the Strait of Gibraltar, flows towards the Spanish coast and curves to the south to form the anticyclonic gyre (WAG). On the other hand, surface water velocity reaches its maximum values (0.55 m/s) in the Strait of Gibraltar which is in good agreement with some measured currents in the same area (0.60 m/s) [2]. Surface currents along the Spanish coast are between 0.1 and 0.4 m/s. On the other hand, the circulation of deep waters is directed towards the west with currents in the Strait of Gibraltar of about 0.15 m/s, also in good agreement with those obtained from Ref. [8]. Thus, the water dynamic is qualitatively and quantitatively reproduced by the model within the studied domain.

2.2. Dispersion of radionuclides

Transport of dissolved radionuclides in the marine environment is governed essentially by advection and diffusion processes. The 3D form of the dispersion equation that gives the time evolution of concentrations in water can be written as follows:

$$\frac{\partial C}{\partial t}+u.\frac{\partial C}{\partial x}+v.\frac{\partial C}{\partial y}+w.\frac{\partial C}{\partial z}$$
$$=\frac{\partial}{\partial x}\left[K_h.\frac{\partial C}{\partial x}\right]+\frac{\partial}{\partial y}\left[K_h.\frac{\partial C}{\partial y}\right]+\frac{\partial}{\partial z}\left[K_v.\frac{\partial C}{\partial z}\right]- \quad .C \qquad (4)$$

where C is the radionuclide concentration in water, λ is the radioactive decay and K_h and K_v are, respectively, the horizontal and vertical diffusion coefficients. K_h is considered the same in both directions x and y (the value taken is 2.0 m^2/s), while K_v is always parameterized as a function of the eddy diffusivity. It is worth noting that although ^{226}Ra is a particle reactive radionuclide [9], interactions with suspended particulate matter (SPM) have been neglected in our dispersion model because of the low SPM concentrations present in the western Alboran Sea and the Strait of Gibraltar. It has been shown experimentally that the uptake of radionuclides increases as the SPM concentration increases [10].

The advective transport is governed by the residual and tidal currents (u, v and w) previously established by the hydrodynamic models and stored in files to be used by the dispersion model. In order to reduce the numerical dispersion, the MSOU second-order scheme (Monotonic Second Order Upstream) is used to solve the advection term [11].

The model has been applied to simulate ^{226}Ra releases from hypothetical phosphate fertilizer industries located on both the Spanish and Moroccan coasts. Fig. 4 shows an example of the time evolution in surface waters of ^{226}Ra concentrations resulting from a single discharge at the Strait of Gibraltar. The total activity discharged is 100 kBq. After 2 d, the patch starts to be introduced in

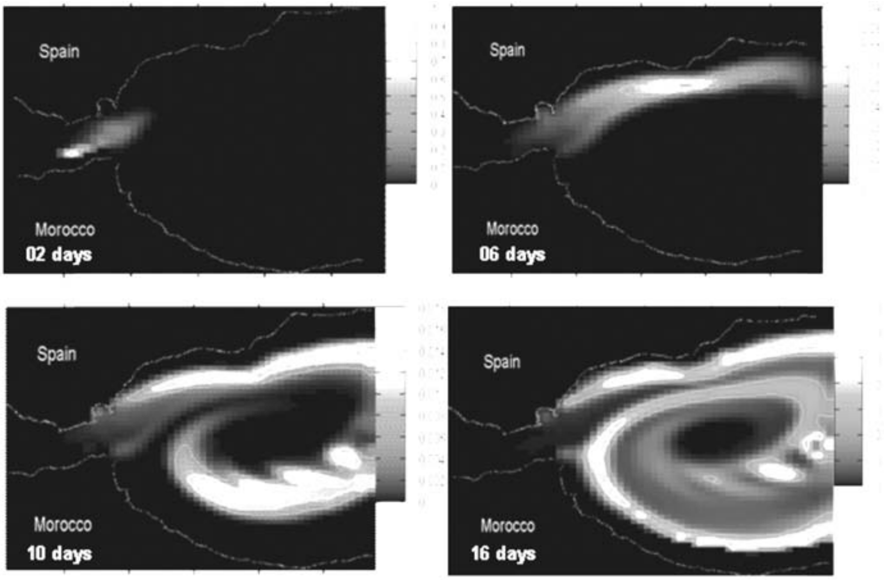

Fig. 4. Spatiotemporal evolution in the surface water layer of ^{226}Ra concentrations resulting from a single discharge at the Strait of Gibraltar [Grid cell (10,27)].

the Alboran Sea due to the Atlantic jet and moves near the Spanish coast. Concentrations decrease as a consequence of dilution with uncontaminated water since the model starts from clean water. Effectively, the concentration decreased from 1000 to 0.1 Bq per volume unit in two weeks. The vertical transport of ^{226}Ra is also responsible for the decrease in surface concentration. The maximum vertical velocity is of the order of 10^{-4} m/s which makes the vertical advective transport significant only for long periods of simulation.

The patch then curves to the south and part of the ^{226}Ra is retained within the Alboran basin following the WAG for a considerable period, diluting progressively until reaching low concentrations at background levels. From the results above, the transport of ^{226}Ra in the area covered by the model is essentially governed by the water dynamics. Several numerical experiments were carried out using different conditions of ^{226}Ra discharges (location, duration, initial conditions) and the dispersion model has shown its consistency in producing reasonable spatiotemporal evolution of concentration within the covered domain.

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RADIOLOGICAL CHARACTERIZATION OF A COPPER–COBALT MINING AND MILLING SITE

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Abstract

Extensive copper and cobalt ore deposits can be found in the Katanga Province of the Democratic Republic of the Congo near the city of Kolwezi. These deposits have been mined via open pit and underground mines since the 19th century. With the recent re-establishment of a relatively stable democratic government in the DRC, foreign investors returned to the area to restart mining activities that were abruptly terminated in the 1990s due to political turmoil. Some of these new projects are being performed in accordance with World Bank and International Finance Corporation Social and Environmental Sustainability standards. As part of these standards, radiological characterization of the mines, processing facilities, and surrounding environment was conducted to establish current conditions, evaluate human health and ecological risks and provide a basis for the establishment of radiation safety and environmental remediation programmes. In addition to naturally occurring radioactive materials associated with the copper/cobalt ore, the site was reputedly historically used to store ore from the Shinkolobwe uranium mine, the source of the uranium ore for the World War II Manhattan Project. The radiological characterization was conducted via extensive gamma radiation surveys, random grid composite soil sampling, biased soil sampling of areas with elevated gamma radiation levels, and sampling of surface water features. The gamma radiation surveys revealed that elevated radiation levels were largely confined to areas previously disturbed by mechanized mining activities. Radiological contaminants in local surface water sources were within drinking water standards with the exception of one river heavily polluted with both uranium and other metals by waste streams from an ore processing and refining facility. Surrounding areas that appeared to be undisturbed by mining, including agricultural areas, native villages, and urban colonial architecture cities, exhibited soil concentration and gamma radiation levels consistent with expected background levels.

1. INTRODUCTION AND BACKGROUND

Prior to the recent economic downturn, copper–cobalt projects were being developed near the city of Kolwezi in the Katanga Province of the Democratic Republic of Congo (DRC). In addition to mining of unprocessed ore, copper and cobalt are being reclaimed through reprocessing of the tailings deposits from previous mining and milling operations.[1] Over 110 million tonnes of ore have been processed in this area during almost a century of mining, primarily for copper and cobalt minerals. As a result, the area around Kolwezi has numerous pits, waste rock, ore and slag dumps, tailings dams, concentrators and other mine-related infrastructure. In addition, the sites for two former uranium stockpiles located in the vicinity of operations represent potential source terms.

Environmental and social studies are required for the compilation of an Environmental Safety Impact Assessment or Environmental Safety Management Plan (ESIA/ESMP) to International Finance Corporation (IFC) Standards. In addition, the Department for the Protection of the Mining Environment in the DRC requested that radiation surveys to characterize the risk of radiation resulting from the mining and processing operations to the workforce and surrounding communities be performed. Preliminary indications were that there was likely to be extensive distribution of radioactive material in the ground and surface water, air and soil, indicating the need to conduct radiological characterization surveys and associated dose and risk assessments to determine the extent and significance of site source terms with respect to human health and the environment.

2. SCOPE OF WORK

The radiation study activities were divided into two phases. The first phase was designed to accomplish preliminary characterization activities sufficient to permit the detailed design of the scope and cost of a second phase of characterization. The second phase of the project will then be implemented to collect the detailed information necessary to perform a full radiological dose and risk assessment. The information collected in this first phase was not intended to be detailed enough to perform such an assessment or provide detailed estimates of costs for a feasibility study. However, in addition to scoping a second phase, a

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[1] In this paper, the term ‘mining’ is used to describe non-artisanal mining activities, that is, mining using industrial, mechanized methods. Artisanal mining activities are referred to specifically as ‘artisanal mining’.

preliminary risk assessment was performed with the available data. These data are sufficient in many cases to determine that there are no radiological hazards associated with given areas, but are not sufficient to definitively characterize the risks in areas containing or potentially containing hazards.

The project was designed to consider and meet the appropriate social and environmental standards including, but not limited to:

(a) DRC law and regulations such as the Mining Code;
(b) IFC Policy and Performance Standards (PS) and associated Guidance Notes;
(c) The Pollution Prevention Handbook;
(d) Sector and industry specific guidelines and standards;
(e) World Bank Environment, Health and Safety guidelines.

During the phase 1 study, a broad-area gamma radiometric survey of the requested areas was performed. Soil samples were collected, as were limited water samples and limited radon emanation measurements. No air sampling was performed and groundwater sampling was limited. Of the samples taken and survey measurements made, the intent was to identify areas where further study was and was not needed rather than to fully characterize the extent and nature of the hazards in each area.

3. METHODOLOGY

Gamma radiation surveys over the entire concession area and portions of adjoining areas were completed. Instruments used were Ludlum Model 2241 series survey meters with a Ludlum model 44-20 7.6 × 7.6 cm (3 × 3 inch) NaI scintillator probes. This type of detection unit is sensitive to low levels of gamma radiation, such as that found in environmental monitoring situations. The probes were positioned at a height of approximately 1 m. The majority of the surveying was performed with the radiation detectors mounted on four-wheel drive vehicles. Vehicle speeds were kept below 30 km/h during surveying and averaged 15–20 km/h. Select areas (tailings areas, waste rock areas, uranium ore storage areas) were surveyed on foot. Only surface levels were measured; surveys were not conducted within artisanal mining pits or holes, or any other subsurface openings. Outside of the concession area, limited gamma survey data was obtained, mostly limited to roads, wide foot and bike paths and similar accessible transit routes; however, surveys outside the concession area and in relatively unimpacted portions of the concession area were sufficient to establish background levels.

Areas within and adjacent to the concession area were divided into 1 km square units. Within each unit, soil at three of five randomly chosen locations was sampled and composited for analysis. Hand held GPS units were used to locate the exact positions. If a position fell within a body of water or other inaccessible or inappropriate location, an alternate location was selected from the 5 random points. In addition, targeted samples from within the concession area were sampled based primarily on elevated gamma survey results at that location. Surface debris and organic material were removed and approximately 2 kg of material was collected from each sampling location by digging out a 15 cm dia × 15 cm deep portion of soil. Samples were collected in sample bags, labeled and sealed.

Radon emanation measurements were made at approximately 10 locations within the concession area. Locations were chosen based upon gamma survey results, accessibility, and ability to hide the measurement devices. (Two sets of instruments were stolen during data collection).

Water samples were selected from surface bodies of water. No samples from monitor wells or domestic or community water supplies other than surface ponds were taken due to a lack of access. Samples upstream and downstream of the concession area were taken where access permitted. Two litres of water were collected from each sampling location using a pair of 1 L polyethylene bottles.

Soil samples were analysed using inductively coupled plasma mass spectrometry (ICP–MS) for total uranium and thorium concentrations and with gamma spectroscopy for gamma-emitting isotopes. Water samples were analysed using ICP–MS for total uranium and thorium concentrations, gamma spectroscopy for gamma-emitting isotopes and radiochemical separation for ^{226}Ra and ^{228}Ra.

4. RELEVANT LEGISLATION, STANDARDS AND GUIDELINES

The DRC does not have a regulatory agency appointed specifically with the task of regulating radioactive material. However, the DRC is a Member State of the IAEA. The IAEA states in its International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Sources (BSS) [1] that its standards are not regulatory law, but are to be used as a practical guide for individual governments as needed. The World Bank and IFC specify that certain standards be applied concerning the safe operation of any project utilizing their funding [2–4]. The BSS and various other IAEA publications [5–8] can be utilized as accepted industry standards. The IAEA standards provide quantitative guidance concerning the assessment of the impacts of radioactive material whereas the IFC standards generically address workplace and environmental hazards but do not provide quantitative guidance concerning acceptable levels of potential contaminants. In addition, the DRC Mining Code contains applicable requirements [9].

There are no specific international standards or recommendations for the regulation of NORM in the environment. However, some IAEA Member States have developed standards that have now become the industry standard. Radioactive material is generally considered outside the scope of regulation if the concentrations are less than 10 Bq/g for ^{40}K, and 1 Bq/g for other radionuclides. Areas deemed to have concentrations above certain values should be restricted access areas. The intent is to protect individual members of the public from receiving doses in excess of 1 mSv/a (or a collective dose of 1 man·Sv/a when considering groups of persons). The IAEA also states that the 1 Bq/g criterion for regulation can be reconsidered on a case by case basis; this might be especially relevant in emerging markets outside of G8 countries. IAEA recommends general safe practices and programme requirements for working with and around radioactive material, consistent with the recommendations of the ICRP in its Publication 60 [10].

The IFC performance standards cover all hazardous materials and are not specific to radiation. The standards outline programmatic features that should be in place to ensure the protection of the public, the environment, the workers and the investment IFC has made in the particular industry [2]. The mining entity must develop an Environmental Management Plan (EMP) wherein the operational safety goals concerning the public, the environment, the workers and the capital investment are described. An environmental action plan must also be written to describe how the exposure limits, effluent concentrations and other goals outlined in the EMP will be met.

The World Bank has developed mining standards which are very similar to the IFC performance standards with regard to general programme features. However, the World Bank standards include safety guidelines specific to radiation for mining industry workers [3]. This includes standards for the development of a safety management plan for radiation hazards, effluent monitoring, hazard characterization and mitigation and proper disposal of hazardous waste. Workers expected to receive more than 6 mSv/a must take part in a dosimetry programme. Dose limits are specified as 20 mSv/a when averaged over five consecutive years, with no single year to exceed 50 mSv.

All mining activity in the DRC falls under the jurisdiction of the Minister of Mines. Local authority is given to the Provincial Mining Registrar to govern operations of mining activities pursuant to the DRC Mining Code [9]. The DRC Mining Code does not contain any specific requirements related to radioactive material and only addresses radioactive material generically under its general hazardous material controls. The Mining Code essentially contains all the elements of the IFC standards along with a chronological order in which all management plans must be developed and put into place. A complete environmental impact statement and EMP are required to be approved before mining activities can begin. If the environment is to be altered, such as emplacement of tailings piles or capping of areas containing radioactive materials, an environmental adjustment plan must also be filed and approved. Radiation hazards and mitigation measures should be specifically included in all environmental plans filed with the DRC government. The storage of mineral products cannot endanger the environment or the public.

A worker safety and industrial hygiene programme must be put into place and should specifically address the formation of a radiation safety programme. The DRC Mining Code also includes provisions for inspections by government officials. It also holds mining entities liable for damages and financial compensation should an environmentally or socially negative impact be caused as a result of mining operations.

5. RESULTS

A total of 109 soil samples were submitted for analysis. Of these, 25 were ‘biased’ or ‘targeted’ samples collected from identified areas of elevated radiation levels in the Kolwezi concession area. The remaining samples included five duplicates (approximately 1 in 20) and were composite samples collected from within and around the concession areas. Twenty-two water samples were collected.

The results of the gamma radiation surveys indicate that the majority of the Kolwezi concession area and the immediate surroundings are consistent with expected natural background radiation levels of 0.1–0.2 µSv/h or less, as shown in bright green in Fig. 1. Some areas directly impacted by mining activities exhibited elevated radiation dose rates. There were several locations with substantially elevated radiation levels:

(a) Both uranium ore storage locations;
(b) The copper refinery tailings area;
(c) The copper concentrator ore concentrate storage area;
(d) Limited areas on waste rock piles.

In addition to these areas, multiple locations were noted throughout the concession area on roads maintained by the mining company that had small, discrete spots with slightly elevated readings. These appeared to be associated with a white, chalky material that formed a part of the road base. All of these

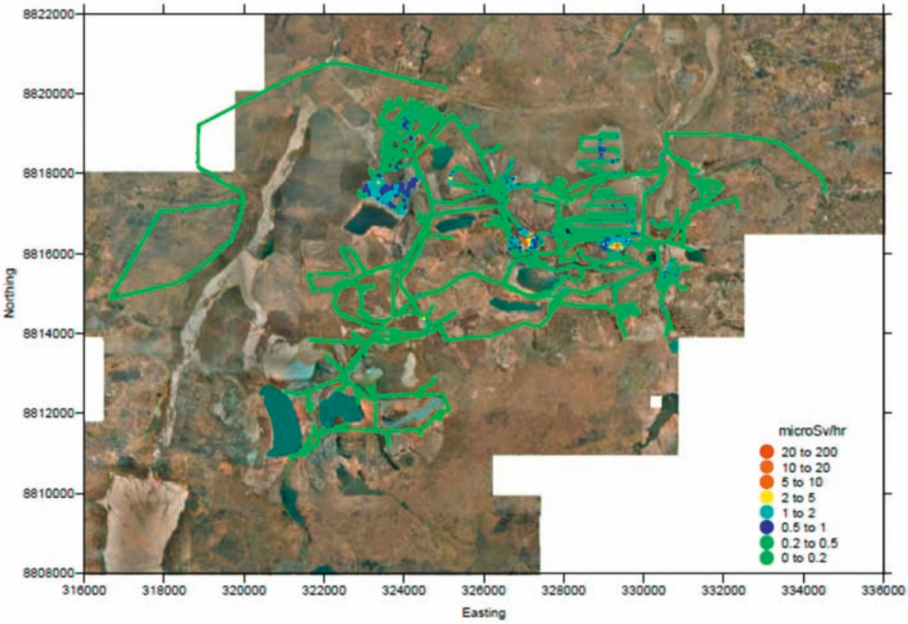

FIG. 1. Kolwezi gamma survey.

areas exhibited radiation dose rates of 1 µSv/h or greater up to a maximum observed dose rate of approximately 160 µSv/h.

In addition to the substantially elevated areas, there were several moderately elevated areas that exhibited dose rates of 0.2–0.5 µSv/h with small isolated areas indicating higher levels including:

(1) Tailings areas;
(2) Waste rock dumps;
(3) One open mine pit (of more than nine in the area);
(4) The copper refinery.

The composite sampling performed inside and outside the concession area exhibited similar results to the gamma radiation surveys, with the majority of the results less than 1 Bq/g. In fact, no composite samples had a concentration for ^{226}Ra, ^{228}Ra, ^{238}U, or ^{232}Th in excess of 0.2 Bq/g and thus the summed concentrations were less than 1 Bq/g as well. Figure 2 depicts the concentrations for ^{238}U. The concentrations for the other radionuclides were similar or lower in magnitude. With few exceptions, the soil samples exhibited a good correlation between the concentrations of ^{238}U and ^{226}Ra and between ^{232}Th and ^{228}Ra. The ^{232}Th concentrations were equal to or less than that of the ^{238}U. In areas with

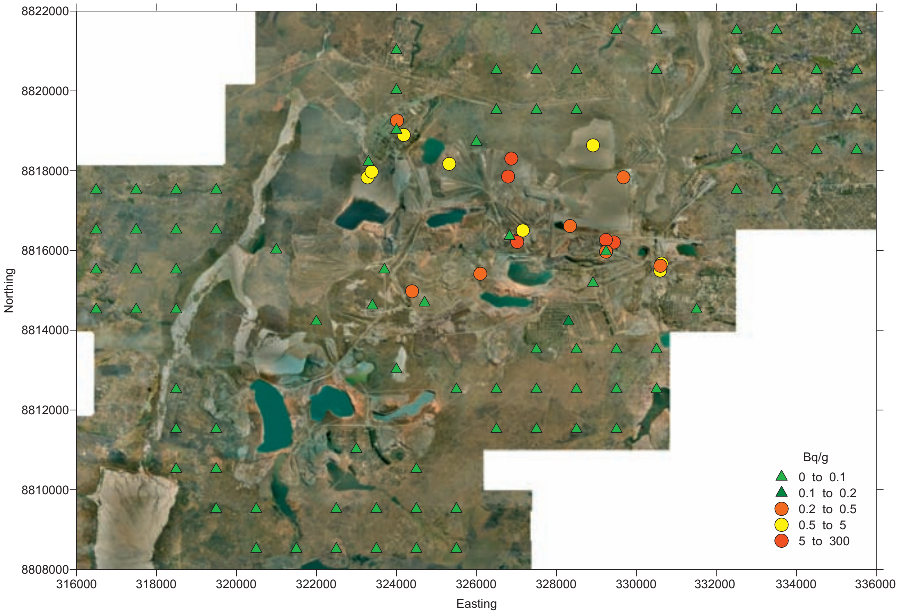

FIG. 2. Kolwezi ^{238}U concentrations.

elevated ^{238}U concentrations, the ^{232}Th concentrations were generally much less elevated.

A few water samples were taken, including seven from mine pits, three from lakes, three from ponds on inactive tailings areas, seven from surface streams and rivers, and two from waste water streams associated with the copper refinery. With respect to NORM concentrations in water, US EPA standards for municipal water supplies were used to establish exemption levels. Concentrations below those levels were deemed to have no adverse impact. These levels are 0.185 Bq/L of ^{226}Ra and ^{228}Ra combined and 30 µg/L of uranium. Of the water samples collected, six samples exceeded the 30 µg/L uranium concentration exemption level, of which only one was associated with a natural water body, the Luilu river downstream of the copper refinery. Levels exceeding the 0.185 Bq/g combined ^{226}Ra–^{228}Ra level were observed in three water samples, with the highest being a seasonal pond on an inactive tailings pile.

5.1. Uranium ore storage locations

Two locations within the concession area were observed to be posted with signs warning people about the presence of uranium, radiation and radioactivity. The radiation dose rates in these areas ranged from 2 to 160 µSv/h. The external dose rates in this area are sufficiently high to exceed recommended dose limits for members of the public in a very short period of time. While the majority of the material in this area is relatively rocky, there is sufficient finer grained material that may become airborne to potentially pose an inhalation hazard. The overall size of these areas is limited, especially with respect to the overall site, limiting the impact of airborne materials at a greater distance. The gamma radiation surveys do not indicate extensive airborne dispersion of material from these areas.

5.2. Copper refinery tailings area

South of the copper refinery is what appears to be a semi-active tailings area. The accessible (mostly dry) portions exhibited elevated radiation levels as well as elevated levels of uranium and uranium decay products. Although stable enough to walk on, the tailings appeared to be quite saturated and thus are not likely to pose an inhalation hazard, although incidental ingestion remains a possibility. The external dose rates in this area are sufficiently high to exceed recommended dose limits for members of the public.

5.3. Copper concentrator

Elevated radiation levels, as well as elevated levels of uranium and uranium decay products, were observed in the western portion of the facility where ore concentrate was being stored in ‘supersacks’. The elevated radiation dose rates appeared to be directly correlated with the presence and thickness of a layer of powdery grey material on the ground. This material was notably drier and lighter in colour than the ore concentrate piles. Due to the fine particle size of this material, it poses an inhalation hazard as well as an external dose hazard. Some of the workers moving the ore concentrate supersacks were noticed to be wearing surgical mask type respiratory protection. While workers were observed to possess these masks in several locations, this is one of few instances where the masks were actually seen in use.

5.4. Waste rock piles

The radiation levels at the waste rock piles varied from levels consistent with background to substantially elevated rates. The indications are that the majority of the rock piles are at background levels with only moderately elevated readings elsewhere. Small elevated areas were observed to be associated with localized lower elevation areas where sediments had collected from rain runoff. Whether the elevated readings are associated with the collected sediments or are a result of uncovered underlying materials is not known at this time. One substantially elevated region is larger and not directly associated with lower elevation areas. The reason for these elevated readings is unknown. A possible explanation is that materials from one of the uranium ore storage areas was transported to the waste rock pile along with overburden materials from one of the open mine pits.

5.5. Tailings areas

The majority of the tailings areas exhibit only moderately elevated readings, between 0.2 and 0.5 µSv/h, corresponding to approximately 0.05–0.35 µSv/h above background levels. Limited areas exhibited dose rates in the 0.5–1 µSv/h range. There were qualitative indications that the more elevated areas were associated with hard-pack areas or where metal debris was located. It is probable that these tailings areas have been previously used as disposal areas for process equipment, which may account for the localized elevated dose rates.

Although the external dose rates alone are not sufficient to pose an issue, the fine grained nature of the tailings does indicate the potential for an inhalation hazard. Evidence of wind-blown tailings was noted on all sides of the tailings areas and especially the east sides. The ^{238}U and decay product concentrations in the most of the tailings are below the IAEA regulatory criterion of 1 Bq/g, which was treated here as an exemption level. However, the ^{238}U and decay product concentrations in the sulphide ore tailings exceed this level with sampled concentrations of 1.9 Bq/g ^{238}U and 1.7 Bq/g ^{226}Ra. Evaluation of inhalation hazards for these areas should be addressed. As the largest potential source of airborne dust and obvious indication of airborne transport, evaluation of the inhalation hazard of these areas should be prioritized.

5.6. Open mine pits

The areas around two of the open pits exhibited moderately elevated radiation dose rates of 0.2–0.5 µSv/h. In these areas, more surveys were performed at elevations below what was the normal ground surface before mining commenced. These elevated readings may be the result of increased uranium concentrations associated with more highly mineralized ore zones. Regardless, the increase is not sufficient to exceed dose limits for members of the public. The results of the one composite sample collected from these areas did not indicate any activity concentrations exceeding the exemption level of 1 Bq/g. Therefore, the potential for inhalation or ingestion hazards associated with these areas are minimal.

5.7. Copper refinery

Most of the surveyed portions of the Luilu plant exhibited moderately elevated radiation dose rates of 0.2–0.5 µSv/h. However, the surveys were primarily limited to the roadways. It must be noted that the interiors of the buildings were not surveyed. Given the radiation dose rates and radionuclide concentrations in the associated tailings area, investigation of the interior of the ore processing buildings and structures is warranted.

Isolated areas with elevated readings were noted in locations where coal furnace slag had been spread out as a dust suppressant, including in front of the shipping and receiving warehouse and a nearby area where welding activities were ongoing. This material was conspicuous for its granular nature and dark greyish-black colour. NORM radionuclides are a known contaminant in coal and are concentrated in the slag during the combustion process. Use of this material as a dust suppressant should be managed to ensure that the dust-suppression advantages of this material are not offset by increased radiation exposure.

Other larger and more elevated areas were noted near various process buildings along the north-west portion of the plant as well as near the electrowinning plant. Additional information concerning these areas and the processes occurring in the adjacent structures is needed to properly assess the hazards associated with these areas.

5.8. Radon measurements

Radon emanation measurements were made in select locations, including the uranium ore storage locations. The radon measurements made in areas with elevated dose rates resulted in saturation of the measuring devices due to the levels of radon and the ambient radiation levels in those areas. Because of the measured radium concentrations in these areas, they are doubtless a significant source of radon. Mitigation measures which will serve to reduce the external dose rate, such as capping or removal of material or exclusion of individuals will also serve to mitigate any radon exposure. Radon emanation measurements in other locations were consistently less than 0.05 $Bq \cdot m^{-2} \cdot s^{-1}$. Levels of less than 0.74 $Bq \cdot m^{-2} \cdot s^{-1}$ may be considered as negligible. Therefore, consideration of radon as a separate hazard was not pursued further.

5.9. Baseline environment

The true determination of a baseline environment requires either survey and sample data for a location prior to the area being disturbed by mining activity or identification of a surrogate area, usually close by and composed of similar geologic formations, which can be considered not to be impacted by the mining activities. Given the long duration and physical extent of the mining activities in the Kolwezi region, this determination cannot be made definitively. However, the surveys and sampling which have been performed around the perimeter of the concession area are consistent with the typical background levels for an area not impacted by mining activities.

Gamma radiation dose rates were typically 0.1–0.2 µSv/h near Kolwezi, although dose rates as low as 0.05 µSv/h were noted. Concentrations of ^{238}U in soil were 0.01–0.1 Bq/g with the associated decay products being present in equilibrium. Concentrations of ^{232}Th in soil were 0.01–0.06 Bq/g with the associated decay products being present in equilibrium.

6. CONCLUSIONS

The surveys and sampling performed during the first phase of this study were sufficient to demonstrate that there is not a widespread hazard from radioactive material. While there are certainly locations that merit the implementation of controls and mitigative measures to reduce the risk associated with those areas, there is no indication of widespread contamination on the perimeter of the Kolwezi concession area or outside. It must be noted that this study did not evaluate groundwater risks and only conducted a limited sampling of surface water sources, nor did it evaluate sources of airborne contamination.

The areas which have been identified as warranting risk mitigation measures are:

(a) Uranium ore storage locations, which require external exposure control;
(b) The copper refinery, which requires external exposure control;
(c) The copper concentrator ore concentrate storage area, which requires external exposure control and dust mitigation;
(d) The tailings area, which requires dust and inhalation hazard mitigation;
(e) The Luilu River.

One of the keys to success will rely on establishing and managing a radiation safety programme that is commensurate with the scope of site activities yet practical to implement, given the challenges of the operating environment. Programmes should be established to provide for external and internal radiation monitoring of personnel; environmental monitoring and control; storage and disposal of radioactive material and waste; contamination monitoring and control; training of personnel; and increasing community awareness of the potential radiation hazards at the site.

Based on the results of the surveying and sampling activities completed to date, a second phase of investigation is warranted to evaluate areas not included in the first phase, or that were identified in that phase as meriting more detailed investigation. Within the mining concession, assessment of the plant and building interiors for the copper concentrators and refinery are advised. Additionally, outdoor tailings areas, waste rock piles and uranium ore storage areas should be evaluated as a sources of airborne dust. With respect to surface water and water supplies used for drinking, additional evaluations of aqueous discharge points, all surface water streams and municipal and residential water supplies should be carried out.

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IDENTIFICATION, HANDLING AND DISPOSAL OF NORM IN THE NORWEGIAN PETROLEUM INDUSTRY

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Abstract

The origin of NORM waste in the form of ‘low specific activty’ (LSA) scale in the oil industry is the formation of sulphate and carbonate precipitates inside tubulars and other production equipment during oil production. The radioactivity is caused by the co-precipitation of small amounts of radium together with the large quantities of barium and strontium. From the gas industry another type of NORM, originating from the decay of radon is known: ^{210}Pb containing iron(II) sulphide. Management of NORM waste can be viewed according to five different waste streams: (i) sediments from production equipment, (ii) scale from production equipment, (iii) suspended NORM, (iv) NORM deposits in gas pipelines and (v) re-injection of NORM. After identification, the NORM is removed from the production and processing sites. NORM contaminated production equipment such as production tubulars, valves and bends is decontaminated, usually by use of high pressure water jetting. Depending on the activity concentration of the NORM waste, disposal is carried out at a hazardous waste disposal site licensed for NORM disposal below the repository acceptance level or at the NORM disposal site at Stangeneset, Sogn og Fjordane County, Norway.

1. PRESENCE OF NORM IN THE OIL AND GAS INDUSTRY

During oil production, NORM is often formed on the interior surfaces of oil production equipment such as tubulars, ‘Christmas trees’, valves and bends in a process where radium co-precipitates with barium and strontium to form hard scales of sulphate or carbonate on the inner steel surfaces. Carbonate scale is later easily dissolved owing to its acidic properties, while the sulphates remain insoluble. The activity concentration of the NORM scale is dependent on the contribution of dissolved radium from the rock in the oil reservoir and the properties and amounts of water associated with the oil in the reservoir (‘produced water’). Aboard an offshore oil-producing installation, NORM may be present in the production equipment prior to the oil–water separation, in the oil–water separation equipment and the water discharge system.

During gas production, radon is generally present, dissolved in the gas. When radon decays (with a half-life 3.8 d), ^{210}Pb is formed and deposited on the inner surfaces of the production equipment and pipelines. Two principal types of this scale are known: (i) the so-called ‘lead scale’ which comprises ^{210}Pb atoms directly assimilated into the steel itself and (ii) an iron(II) sulphide precipitate which is a corrosion product often referred to as ‘black powder’. From this, the NORM waste often occurs as a waste from ‘pigging’ (cleaning) of gas transport pipelines. When this corrosion product is found in oil production equipment, it can be identified by virtue of the fact that the ^{210}Pb activity concentration is substantially higher than the ^{226}Ra activity concentration. The lead scale is invisible and can only be identified through measurements with equipment sensitive to alpha and beta radiation. So far, this invisible lead scale has not been identified at Norwegian gas producing installations. A summary of the various types of scale is given in Table 1.

In a survey by Norse Decom AS in 2003, the mechanical, chemical and radiological composition of LSA scale, comprising more than 90% of the total amount of NORM stored at that time, was determined. More than 130 samples were collected and analysed with respect to physical composition and content of major radionuclides (^{226}Ra, ^{228}Ra and ^{210}Pb). The results, shown in Table 2, showed that the major constituents in Norwegian petroleum NORM waste, disregarding the water content, are sulphate, sand and clay, heavy oil components and corrosion products (rust and steel particles). All stored NORM had been classified according to the clearance level in effect at that time of 10 Bq/g for ^{226}Ra, ^{228}Ra and ^{210}Pb. Most of the stored NORM investigated in the survey originated from decontamination of used production tubulars.

TABLE 1. TYPES OF RADIOACTIVE SCALE IN THE PETROLEUM INDUSTRY

	Main constituent	Main radionuclides	Production type
Sulphate scale	Ba/Sr sulphate	^{226}Ra, ^{228}Ra	Oil
Carbonate scale	Ca carbonate	^{226}Ra, ^{228}Ra	Oil
Lead scale	Steel	^{210}Pb	Gas
Sulphide scale (‘black powder’)	Iron sulphide	^{226}Ra, ^{228}Ra, ^{210}Pb	Oil and gas

TABLE 2. MAIN CONSTITUENTS OF STORED LSA SCALE FROM THE NORWEGIAN NORTH SEA SECTOR

	Mass (t)	Composition (wt%)				
		Water	Heavy oil components	Sulphate	Corrosion products	Sand and clay
Company A	166	23.6	7.4	45.7	8.5	14.8
Company B	4.1	15.9	1.4	77.9	2.0	2.8
Company C	0.5	11.8	1.5	75.4	6.8	4.5
Company D	17.0	45.4	6.6	39.0	6.1	2.9

In 2009 a new study on petroleum industry NORM was conducted [1]. The study was funded by the Norwegian Radiation Protection Authorities and carried out by Norse Decom AS, Norway in cooperation with FUGRO-HGN GmbH, Germany.

2. PETROLEUM INDUSTRY NORM WASTE STREAMS

For the petroleum industry, the produced NORM represents a waste that needs to be addressed properly to prevent harm to workers or the environment. The NORM waste occurs as a result of normal maintenance operations such as the replacement of used production equipment or removal of sediments from vessels. To better understand the different aspects and tasks involved in waste handling and processing, the waste can be assigned to the following NORM waste streams:

(a) *Sediments from production equipment.* These comprise NORM contaminated sediments that accumulate in vessels, sand collectors and drains. At intervals (typically during maintenance periods) the sediments are removed from the vessels and drains and brought onshore for disposal.

(b) *Scale from production equipment.* Sulphate scale on the inner surfaces of the production equipment is a well-known source of NORM waste. The waste is produced when the NORM contaminated equipment is decontaminated.

(c) *Suspended NORM.* After processing, the oil can still hold limited amounts of suspended material. If so, there is a possibility for sedimentation of material which can be NORM containing in transport tankers or after offloading at the oil terminal. At present, few data exist on this waste stream and it is not further discussed in this study.

(d) *NORM deposits in gas pipelines.* Owing to the decay of radon dissolved in the produced gas stream that is transported to shore, ^{210}Pb may be deposited onto the inner surface of the gas pipeline.

(e) *Offshore re-injection of NORM.* Instead of being sent ashore, NORM waste from (i) or (ii) can be re-injected into the formation.

NORM contaminated sediments found in offshore production installations are either re-injected (waste stream WS-5 in Fig. 1) or packed and sent ashore (waste stream WS-2 in Fig. 1). Re-injection requires that the waste be slurrified and that there is a suitable well available to receive the material. The process of slurrification requires the use of equipment normally used for processing and pumping of drill mud and cuttings. Depending on the amounts of material involved, containers used for shipment of material to shore vary in size from drums (110 L and 220 L) to waste containers and tanks up to 10 m^3.

It is often difficult to perform representative measurements or sampling on site. Logistic factors like availability of deck space and time constraints limit the possibilities for sending samples ashore for analysis at external laboratories. Assessments of NORM at offshore installations are therefore usually performed by means of measurements using hand held equipment such as contamination monitors and dose rate meters.

The activity concentration of NORM contaminated sediments from oil and gas production is usually low, reflecting the fact that this is a mixed waste where the NORM scale component is diluted with sand, clay and heavy oil components. A distribution of ^{226}Ra activity concentrations for 198 sediment samples analysed at Norse Decom’s laboratory is given in Fig. 2 showing a left skewed distribution where most of the samples have a ^{226}Ra activity concentration below 5 Bq/g. It should be noted that the data set is probably biased due to the fact that a large number of samples with low activity concentrations are classified as non-radioactive and are therefore not included, giving a massive under-representation of samples at low activity concentrations.

NORM contaminated production equipment can be decontaminated aboard the installations, but usually it is more convenient and less expensive to dismantle it and send it ashore for decontamination (waste stream WS-1 in Fig. 1). Most of the contaminated equipment consists of production tubing. A typical tube string may be 3000 m long (250 tubes) and can contain several tonnes of NORM scale with a ^{226}Ra activity concentration of 20–100 Bq/g. Other types of contaminated equipment may be valves and bends from the pre-separation lines and the produced water system. The decontaminated equipment is either scrapped or

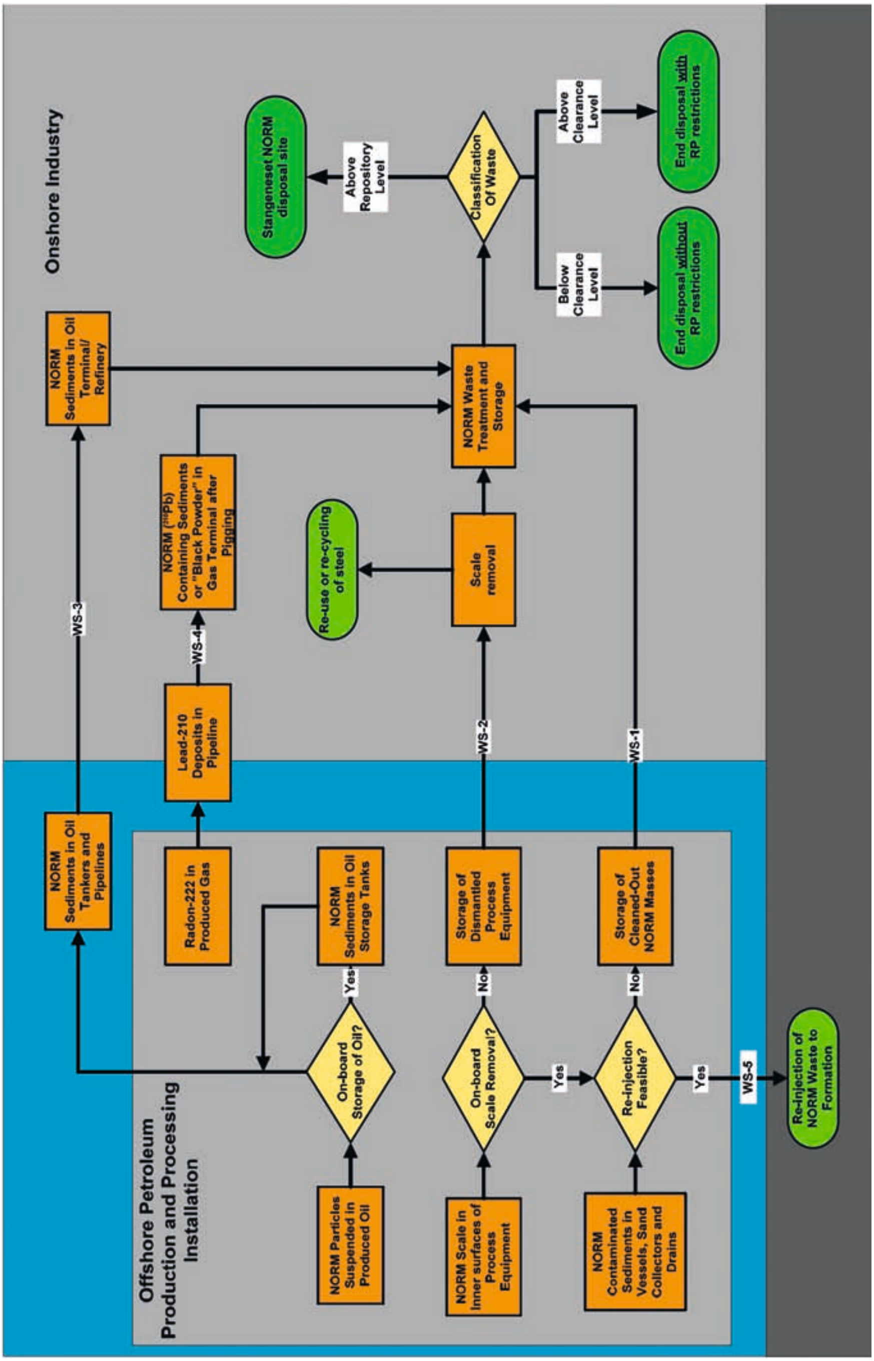


FIG. 1. NORM waste streams.

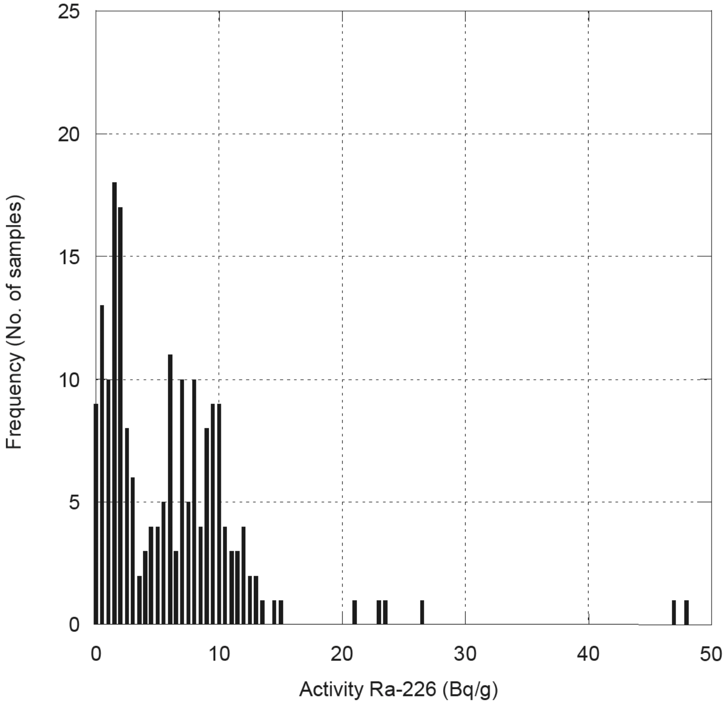


FIG. 2. Distribution of ^{226}Ra activity concentrations in 198 sediment samples, truncated at 50 Bq/g (two samples had an activity concentration >50 Bq/g).

reused. The NORM waste is temporarily stored by the decontamination vendor awaiting final disposal.

The formation of residues containing ^{210}Pb in petroleum gas lines, often denoted as ‘black powder’, is a result of anaerobic corrosion processes on the inner surfaces of the pipeline. The main constituent is iron(II) sulphide. The NORM waste occurs onshore as a solid waste from pigging operations or collected by other means from filters and drains (waste stream WS-4 in Fig. 1). This NORM waste is chemically unstable and should be stabilized before long term storage or disposal.

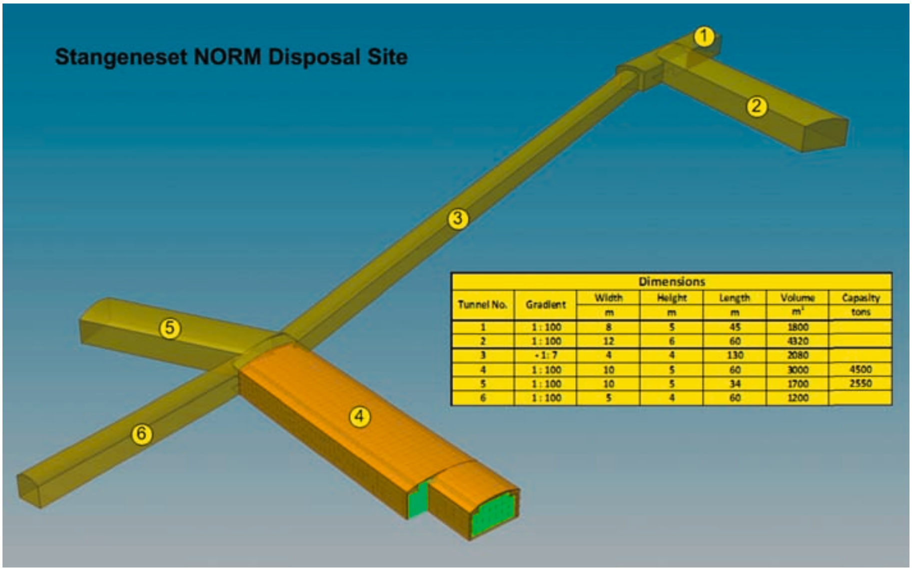
3. NORM WASTE TREATMENT AND STORAGE

NORM contaminated equipment is usually decontaminated using high pressure water-jetting at pressures often exceeding 200 MPa. The NORM scale is collected in sedimentation basins, retrieved, packed in HDPE drums and stored in locked steel containers. The storage facilities are managed by the decontamination companies creating the waste, which are licensed by the Norwegian Radiation Protection Authority. NORM containing sediments above the repository acceptance level (‘repository level’) are packed and stored similarly.

Until now, NORM waste in Norway has been classified only according to the repository level, meaning that this level has also been used as a clearance level. At present (March 2010) the repository level is 5 Bq/g for ^{226}Ra, ^{228}Ra and ^{210}Pb. With the implementation of the proposed clearance level of 0.5 Bq/g, the waste industry is currently working towards treating waste from the petroleum industry with activity between the clearance level and repository level as NORM waste and adapting the methods for its handling and disposal accordingly.

4. DISPOSAL

Depending on the activity concentration of the NORM waste, disposal is either done at a hazardous waste disposal site licensed for NORM disposal below the repository level or at the NORM disposal site at Stangeneset, Sogn og Fjordane County (see Fig. 3). Disposal of NORM with an activity concentration below the repository level is carried out under a licence from the Norwegian Radiation Protection Authority in the same way as non-radioactive waste but with



Dimensions						
Tunnel No.	Gradient	Width	Height	Length	Volume	Capasity
		m	m	m	m^3	tons
1	1 : 100	8	5	45	1800	
2	1 : 100	12	6	60	4320	
3	-1 : 7	4	4	130	2080	
4	1 : 100	10	5	60	3000	4500
5	1 : 100	10	5	34	1700	2550
6	1 : 100	5	4	60	1200	

FIG. 3: Stangeneset NORM disposal facility.

the addition of some degree of basic radiation protection and radiological monitoring. The NORM disposal site at Stangeneset was opened in October 2008 after eight years of planning and preparation. The facility is constructed as a near surface rock cavern repository also containing infrastructure for short term storage and conditioning of the NORM waste packages (see Fig. 3). To date, 200 t of NORM waste have been received and disposed of at Stangeneset.

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USE OF NORM PRODUCTS AND RESIDUES

(Topical Session 5)

Chairpersons

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FLUXES OF ^{238}U SERIES RADIONUCLIDES IN THE INDUSTRIAL PRODUCTION OF DICALCIUM PHOSPHATE AND THE BIOKINETIC ANALYSIS OF ^{210}PB AND ^{210}PO IN POULTRY TISSUE DUE TO ITS INGESTION

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Abstract

NORM derived from the wet acid digestion of phosphate rock might represent a serious problem facing the phosphate industry. The aim of this work is to discuss both: (a) the fluxes of the ^{238}U series radionuclides (U and Th isotopes, ^{226}Ra, ^{210}Pb and ^{210}Po) during the DCP production process and, (b) the accumulation of ^{210}Pb and ^{210}Po in chicken tissues due to the ingestion of DCP. Industrial samples were collected from different production steps at a factory in Spain that uses phosphate rock from Morocco. The results showed that whereas uranium is equally distributed within products and by-products, thorium and ^{210}Po are mainly eliminated through the sludges and ^{210}Pb is mostly found in the final product (DCP). The ^{210}Pb and ^{210}Po results in chicken tissues and faeces after animals were bred with different amounts of DCP for 42 days have shown that about 95% of the ingested ^{210}Pb and ^{210}Po was immediately excreted. From the amount accumulated in tissues it was observed that whereas ^{210}Pb is mostly found in bones, ^{210}Po accumulates in the liver and kidneys. A one compartment non-linear kinetic model has been developed in order to determine the variation of the total activity of ^{210}Pb and ^{210}Po accumulated in the chicken body throughout its 6 weeks life. This model correctly reproduces experimental results and also allows calculating of transfer rates for ^{210}Pb and ^{210}Po, useful for first order models, that is, under stationary metabolic conditions.

1. INTRODUCTION

Phosphate rock is the raw material used for the production of the feed supplement dicalcium phosphate (DCP), which is a supply of calcium for domestic animals such as poultry and cattle [1]. Owing to the replacement of calcium by uranium in the apatite structure during the formation of sedimentary phosphate rock, ^{238}U and its progeny are present in this geological formation at activity concentrations that may exceed 10 Bq/g [2]. During the industrial process, phosphate rock is initially digested with either sulphuric acid (H_2SO_4) or hydrochloric acid (HCl). The former leads to phosphoric acid as the main product and phosphogypsum as a by-product [3], whereas the latter is used to obtain DCP directly as a final product [4]. It has been observed that the enhancement of radionuclide concentrations in products (DCP) and by-products (mud and residual water) strongly depends on the acid used for the rock digestion — whereas the digestion of the rock with H_2SO_4 leads to elevated concentrations of ^{238}U and ^{230}Th in DCP, digestion with HCl leads to elevated activity concentrations of ^{210}Pb and, with time, ^{210}Po [5].

The concern of having a product with activity concentrations of ^{210}Pb and ^{210}Po as high as 2 Bq/g is that it might contribute to internal dose in humans if ingestion and accumulation in domestic animals' meat were significant. Indeed, biokinetic models for ^{210}Pb and ^{210}Po in humans have been developed in recent decades [6, 7], as their intake might result in subtle and adverse health effects. So far, however, biokinetic studies of radionuclides have been mainly focused on laboratory animals to further extrapolate the data to the human body [7, 8] and few data have been published regarding radionuclide accumulation in food producing animals [5, 9], which may further be introduced in the human food chain. A way of illustration is through the study of the production of chickens, turkeys and ducks, which is undergoing expansion due to the very fast growth rates achievable, the high feed conversion ratio and low levels of activity [10]. Therefore, an understanding of the factors affecting the transfer of radionuclides to, and their behaviour in animals is also essential to predict activity concentrations in animal derived food products and to develop effective and appropriate countermeasures [11].

The aims of the present work were therefore to (i) elucidate the fluxes of radionuclides in the ^{238}U decay series in the production process of DCP; (ii) examine the accumulation of ^{210}Pb and ^{210}Po in chicken tissues during its growth as a function of the type and amount of DCP in chicken diets as well as the content of radionuclides; and (iii) build a suitable kinetic model to understand the distribution of ^{210}Pb and ^{210}Po within chicken tissue after ingestion.

2. MATERIALS AND METHODS

2.1. Industrial samples

Several samples were collected at different stages of the production process at a factory located in the north-east of Spain, which produces DCP. The main inputs to the system are phosphate rock (sample S1) and calcium carbonate (sample S5), while the outputs are DCP (sample S10), waste in the form of sludge (sample S9) and liquid effluent (samples S7 and S8). Sample S3 is the non-digested rock that is either recycled to the reactors or discharged as sludge; whereas sample S4 represents the digested fraction of the phosphate rock that leads to the DCP production. A description of the samples is provided in Table 1. Sampling was performed on three occasions: May 2007, November 2007 and April 2008.

2.2. Experimental setup with chickens

Three commercial diets based on corn and soya bean were prepared using two different types of DCP. Diet A, set as the blank diet, contained 2.5% tricalcium phosphate (TCP) with initially low activity concentrations of ^{210}Pb and ^{210}Po (0.0025 ± 0.0002 and 0.0025 ± 0.001 Bq/g, respectively). Diets B and C contained 2.5% and 5% of DCP, respectively. Diet B had 0.056 ± 0.005 Bq/g of ^{210}Pb and 0.062 ± 0.004 Bq/g of ^{210}Po, while diet C had 0.109 ± 0.013 and 0.116 ± 0.006 Bq/g of ^{210}Pb and ^{210}Po, respectively.

The first experiment was carried out in November 2006, in which chickens were fed until the 21st day of life (first age). A second experiment was carried out in June 2007, when chickens were fed until the 42nd day of life (second age). Chicks were reared until the 7th day with a standard diet (containing TCP from the blank diet) and on day 7 they were split up according to the different diets (A, B and C).

Five animals were fed with each diet for the first age experiment: three were slaughtered for the analysis of the whole animal and, for the other two, different tissues (thigh, breast, bone, kidney and liver) were analysed. The second age experiment included 15 individuals per diet: nine of them analysed entirely and six of them slaughtered to analyse the tissues. The weight and amount of food ingestion of each animal was controlled weekly during the experiment. As expected, the growth performance did not differ among experimental diets. Samples of thigh, breast, bones, kidney and liver as well as of the whole animals were obtained after 21 and 42 d. Samples of faeces were collected at weeks 1, 2, 3 and 5. The samples were pooled per replicate and freeze-dried before analysis.

TABLE 1. ACTIVITY CONCENTRATIONS IN SAMPLES FROM THE PRODUCTION OF DCP

Sample	Value	Activity concentration (Bq/g except where stated otherwise)				
		^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po
S1: Phosphate rock	Min.	1.648 ± 0.125	1.601 ± 0.109	1.486 ± 0.100	1.808 ± 0.165	1.748 ± 0.045
	Max.	1.815 ± 0.061	1.959 ± 0.127	1.968 ± 0.044	1.932 ± 0.120	1.949 ± 0.256
S2: Phosphate rock pulp	Min.	1.615 ± 0.032	1.715 ± 0.095	1.489 ± 0.056	1.751 ± 0.057	1.632 ± 0.077
	Max.	1.680 ± 0.088	1.729 ± 0.071	1.751 ± 0.057	1.926 ± 0.136	1.892 ± 0.080
S3: Undigested phosphate rock	Min.	0.563 ± 0.009	0.031 ± 0.002	0.626 ± 0.115	0.488 ± 0.009	0.141 ± 0.029
	Max.	5.369 ± 0.423	1.880 ± 0.175	4.541 ± 0.254	1.727 ± 0.114	9.734 ± 0.639
S4:Digested phosphate rock [a]		nd	nd	nd	75.016 ± 6.802	10.648 ± 0.634
S5: Calcium carbonate		0.054 ± 0.005			0.055 ± 0.002	
S6: Digested rock + calcium carbonate	Min.	0.362 ± 0.011	0.047 ± 0.004	0.214 ± 0.010	1.064 ± 0.022	0.026 ± 0.017
	Max.			1.475 ± 0.046	1.907 ± 0.138	0.087 ± 0.022
S7: Residual water [a]	Min.	nd	nd	43.875 ± 2.281	nd	0.0061 ± 0.0003
	Max.			54.972 ± 2.703		

TABLE 1. ACTIVITY CONCENTRATIONS IN SAMPLES FROM THE PRODUCTION OF DCP (cont.)

Sample	Value	Activity concentration (Bq/g except where stated otherwise)				
		^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po
S8: Liquid plant effluent [a]	Min.	nd	nd	9.104 ± 0.506	0.006 ± 0.002	0.008 ± 0.003
	Max.			19.693 ± 1.068		0.039 ± 0.002
S9: Residual mud	Min.	5.036 ± 0.333	10.090 ± 0.811	0.657 ± 0.042	0.657 ± 0.042	7.945 ± 0.319
	Max.	5.314 ± 0.305	11.905 ± 0.749	2.341 ± 0.187	1.181 ± 0.032	12.983 ± 0.359
S10: DCP	Min.	1.469 ± 0.066	0.063 ± 0.004	0.103 ± 0.004	2.012 ± 0.041	0.102 ± 0.002
	Max.	1.746 ± 0.062	0.393 ± 0.028	0.203 ± 0.005	2.071 ± 0.076	0.188 ± 0.031

[a] Results are expressed in Bq/L.

2.3. Sample pre-treatment and radioisotope analysis

The solid samples were dried at 80ºC and homogenized before analysis. For the analysis of ^{238}U, ^{230}Th and ^{210}Po, about 100–300 mg of sample were digested using a mixture of HCl, HNO_3 and H_2O, after addition of known amounts of internal tracers (^{229}Th, ^{232}U and ^{209}Po). For the radiochemical separation of U and Th, the method described in Ref. [12] was used. The isotopes of U and Th were electrodeposited on a 2.2 cm diameter silver disc in plastic cells using platinum wire as the anode. Lead-210 was auto-plated in a 1M HCl solution onto a 2.2 cm silver disc for 6–7 h at 70ºC with constant stirring. Samples were stored in 125 mL bottles for 1.5 years to determine ^{210}Pb through the measurement of its progeny ^{210}Po. Appropriate decay and ingrowth corrections were applied to determine concentrations at the sampling time [13].

Liquid samples were acidified at pH ~ 1 with concentrated HCl, filtered with a 1 μm Millipore filter and spiked with a known amount of tracer. Pre-concentration of radionuclides in water samples was carried out through iron hydroxide precipitation in a basic medium. Supernatant solutions were discarded and iron hydroxide precipitates centrifuged in a sealed tube and redissolved with 3M HNO_3.

Measurement of the discs was carried out using alpha spectrometry with passivated implanted planar silicon (PIPS) detectors. Ra-226 was analysed by gamma spectrometry through the main line emissions of its progeny ^{214}Pb (295 keV and 352 keV) and ^{214}Bi (609 keV). Between 80 and 100 g of sample were placed into 100 cm^3 geometries, which were sealed and stored for three weeks to ensure secular equilibrium between ^{226}Ra and its short lived progeny. Samples were measured for 1–3 d using a coaxial HPGe detector (GMX, EG&G Ortec). Analyses of the spectra were carried out using Genie 2000 software.

3. RESULTS AND DISCUSSION

3.1. Fate of radionuclides in the DCP production process

The results of the analyses of the samples obtained from the production process of DCP are shown in Table 1, expressed as the range of concentrations measured for the three sampling surveys. It can be observed that concentrations corresponding to phosphate rock (samples S1 and S2), DCP (sample S10) and mud (sample S9) presented little variability. Conversely, samples corresponding to the intermediate steps of the industrial process presented greater dispersion. This may be related to the specific conditions of the production plant at the sampling time. For instance, in May 2007 and April 2008 the plant was not working at its full capacity. In terms of fluxes of radionuclides, it is observed from the same three sampling surveys (see Table 2) that, whereas uranium is equally distributed between the products and by-products, ^{230}Th and ^{210}Po are mainly eliminated through the sludge and ^{210}Pb is mostly found in the final product (DCP).

3.2. Distribution and biokinetic analysis of ^{210}Pb and ^{210}Po in chickens due to ingestion of DCP

The activity concentrations of ^{210}Pb and ^{210}Po in chicken tissue and excrement are shown in Fig. 1, for diet A (2.5% TCP), diet B (2.5% DCP) and diet C (5% DCP). From the amount absorbed in tissues it was observed that, while ^{210}Pb is mostly found in the bone, ^{210}Po accumulates in the liver and kidneys. However, about 95% of the ingested ^{210}Pb and ^{210}Po was immediately excreted through faeces.

A one compartment kinetic model has been developed in order to determine the variation of the total activity of ^{210}Pb and ^{210}Po accumulated in the chicken body throughout the 6 weeks experiment. This model considers: (i) a time-dependent input of ^{210}Pb and ^{210}Po according to the age of the organism; and (ii) two output functions accounting for radioactive decay and for the transfer rate out of the tissues. Since the animals studied were in constant growth during the experiment, steady state conditions could not be considered and thus a non-stationary approach has been developed. The model is derived from a generalization of first order bio-kinetic models [14]:

$$\frac{d}{dt}[x(t)\,p(t)] = -\lambda x(t)\,p(t) - k(t)x(t)\,p(t) + b(t) + x(t)\frac{d}{dt}\,p(t) \qquad (1)$$

TABLE 2: FLUXES OF RADIONUCLIDES IN THE DCP PRODUCTION PROCESS

Sample	Radionuclide flux (Bq per g of DCP produced)				
	^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po
S1: Phosphate rock	2.4–2.6	2.3–2.8	2.1–2.8	2.6–2.8	2.5–2.8
S9: Residual mud	1.5–1.6	3.0–3.6	0.58–0.70	0.20–0.35	2.4–2.9
S10: DCP	1.5–1.7	0.06–0.39	0.10–0.20	2.0–2.1	0.10–0.19

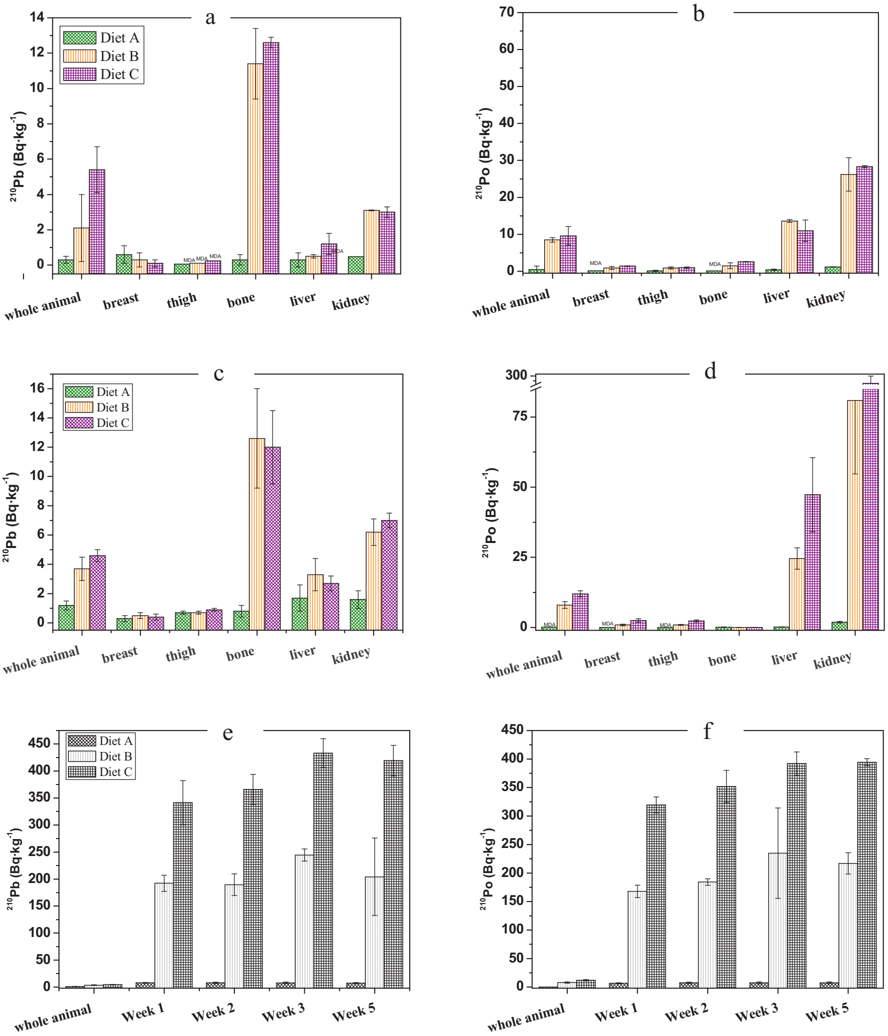

FIG. 1. Activity concentrations of ^{210}Pb and ^{210}Po in tissues after 21 d (a, b) and 42 d (c, d) and in faeces after 1, 2, 3 and 5 weeks (e, f).

where x is the activity concentration of the radionuclide in the whole animal, p is the weight of the whole animal, λ accounts for the radionuclide disintegration, k is the excretion rate that is dependent on metabolism (and thus on time) and b is the radionuclide input through ingestion. The accumulation of radionuclides is shown in Fig. 2 when Eq. 1 is solved and thus non-stationary conditions are considered (solid line). The solution of the standard first order bio-kinetic model

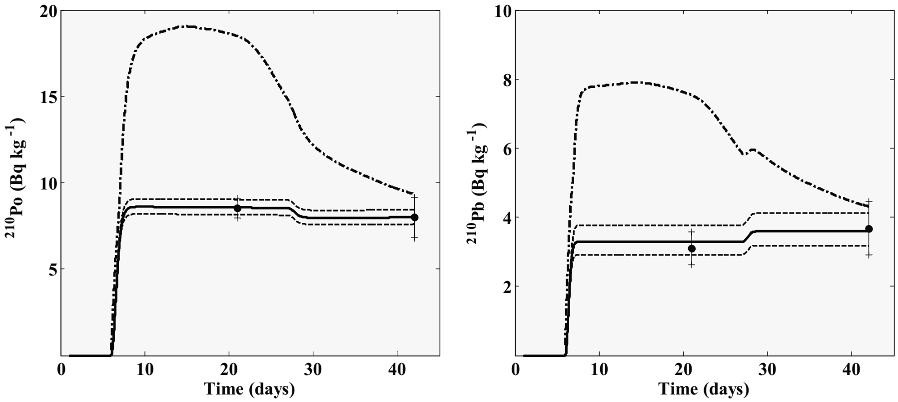

FIG. 2. Activity concentrations of ^{210}Po and ^{210}Pb in the whole animal fed with diet B, as predicted by the first order bio-kinetic model (dot–dashed line) and by Eq. (1) (solid line, plus dashed line representing the uncertainty). The circle symbols represent the experimental results after 21 and 42 days.

(dot–dashed line) is also reported in order to compare the results. Black dots represent the experimental results at days 21 and 42.

This analysis highlights the limitations of the first order kinetic model when strong metabolic variations such as growing occur. The proposed model, as represented by Eq. (1), correctly reproduces the experimental results and also allows the estimation of transfer rates for ^{210}Pb ($k = 3.2 \pm 0.33$ d^{-1}) and ^{210}Po ($k = 1.35 \pm 0.07$ d^{-1}), useful for first order models, that is, under stationary metabolic conditions.

4. 4. CONCLUSIONS

The fate of the main isotopes of the ^{238}U decay series present in phosphate rock has been analysed in different stages of DCP production. The results show that whereas the mud presents high concentrations of ^{238}U (up to 5 Bq/g), ^{230}Th (10 Bq/g) and ^{210}Po (7–12 Bq/g), ^{226}Ra is found in residual waters (~50 Bq/L) and ^{210}Pb and ^{238}U in the final product DCP (2 and 1.5 Bq/g, respectively). In terms of radionuclide fluxes, it is observed that ^{210}Pb is scavenged in the DCP and ^{230}Th and ^{210}Po are eliminated through the mud pathway. Poultry ingesting this DCP might accumulate a small percentage of the total ^{210}Pb and ^{210}Po; however, in previous work we showed that the inclusion of DCP into the human food chain would result in a human dose of <20 μSv/a and therefore poultry ingestion does not represent any significant increase in the dose derived from food consumption.

It is observed that ^{210}Pb accumulates mainly in the bone (up to 0.012 Bq/g), while ^{210}Po accumulates in the liver (about 0.050 Bq/g) and kidneys (up to 0.220 Bq/g). A one compartment kinetic model based on a non-linear approach has been developed; the model correctly reproduces the experimental results and highlights the limits of a first order kinetic model when growing conditions occur.

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RADIOCHEMICAL CHARACTERIZATION OF PHOSPHOGYPSUM FOR ITS USE IN AGRICULTURE IN CERRADO, BRAZIL

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Abstract

This study involved phosphogypsum (PG), a NORM by-product from the so-called ‘wet process’ for the manufacture of phosphate fertilizer. Since it contains U and Th series radionuclides, P_2O_5 and trace metals, it is necessary to understand the mechanisms of radionuclide transfer and to evaluate whether PG in soil contributes significantly to human exposure to natural sources. In greenhouse experiments, PG, soil, soil treated with PG (for lettuce, corn and soya bean cultivation), as well as leached water samples, were analysed for key radionuclides (^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po). The average ^{226}Ra activity in PG (0.252 ± 0.026 Bq/g) was below the limit of the US Environmental Protection Agency. The values of ^{228}Ra, ^{210}Pb, and ^{210}Po were within an order of magnitude of the ^{226}Ra value. Transfer factors varied from 0.00094 to 0.34. In general, the radionuclide mobility in soil was low and demonstrated the viability of PG as a soil amendment in the agriculture of the Cerrado region.

1. INTRODUCTION

The main raw material in the Brazilian phosphate fertilizer industry is the apatite present in rocks, of which approximately 80% are of igneous origin [1]. The most commonly used process in the production of phosphate fertilizer is the attack of the phosphate rock with concentrated sulphuric acid and water. In this case, the main products from the chemical reactions are phosphoric acid (H_3PO_4), simple super-phosphate (SSP), and triple super-phosphate (TSP). Dehydrated calcium sulphate (phosphogypsum) (PG) and hydrofluoric acid are by-products of phosphate rock processing.

Approximately 4.8 t of PG are generated per tonne of phosphoric acid produced. The annual world production is estimated to be 150 million t, approximately 12 million t of which are generated in Brazil [2]. At present, this material is stored in piles located near the fertilizer plants. However, this practice may represent a potential risk of contamination, mainly to organisms and hydrological systems located close to the pile.

The possibility of using PG in agriculture has been the focus of research in several countries [3, 4] and in Brazil, especially in the agriculture of the Cerrado region [5], where soils have characteristics that are compatible with the use of this material. In this system, fast mineralization of organic matter associated with intense leaching produces soils with naturally low fertility. These are acidic soils (pH between 4.3 and 6.2) with high levels of exchangeable aluminum and low levels of phosphorus available for plants; they are also poor in calcium and magnesium, elements involved in root development.

Various residues generated by industries have been studied as possible substitutes for limestone. Among them, PG, or ‘agricultural gypsum’ (AG), has been considered mainly for application in conjunction with limestone in order to improve the effects of the subsuperficial acidity on the root growth. In Brazil, the application of PG as a soil conditioner is a practice that has been carried out for several years [6]. Nevertheless, due to the presence of toxic elements (as an example, heavy metals, metalloids and radionuclides) in PG and taking into account the environmental aspects related to the use of residues in agriculture, a study has been carried out to evaluate whether its application to improve soil fertility can create an impact on human health and the environment.

The migration and accumulation of contaminants in cultivated soils is complex, involving processes such as leaching, capillary rise, runoff, sorption, root uptake and resuspension into the atmosphere. Assessment models normally make use of plant or substrate concentration ratios (TFs) to estimate the transport of radionuclides and other elements to the food [7]. The TF represents the amount of element expected to enter a plant from its substrate under equilibrium conditions. TF values can be influenced by parameters such as soil characteristics, weather conditions, plant type, the plant parts concerned, the radionuclide physico-chemical form and the effect of competitive species [8].

Due to the presence of radionuclides in PG and taking into account the environmental aspects related to the use of residues in agriculture, this study was carried out to evaluate whether its application to improve soil fertility in the Cerrado region of Brazil can result in a radiological impact on human health and the environment.

2. MATERIALS AND METHODS

PG (from storage piles) and soil were radiochemically characterized. Their mixture was employed in several compositions to cultivate lettuce (chosen due to the market demand), corn and soya bean (representative of the Cerrado region). In addition to the cultures, leached water was also radiochemically characterized. Transfer factors in the soil plant system were determined and the dose received by humans from food ingestion was assessed.

2.1. Collection and sample preparation

Samples of PG were collected in a fertilizer facility that produces phosphoric acid by means of the so-called ‘wet process’. The phosphate rock used at the industrial site is of igneous origin and comes from the alkaline-carbonate site in Tapira, MG, Brazil [1]. Thirty samples were collected from the surface of the piles at different locations, according to guidelines of the US Environmental Protection Agency [9]. PG samples were then dried in the laboratory at 60ºC for 48 h and sieved through 30 and 60 mesh (590 and 250 µm) screens. Afterwards, small fractions of the samples were mixed and then divided to form a composite sample.

Soil samples were collected from two different locations in the Sete Lagoas municipal district (clayey yellow rhodic ferralsol (hapludox) — LVSL) and in the Três Marias municipal district (sandy rhodic ferralsol (hapludox) — LVTM), that represent typical soils from the Cerrado region. Soil samples were air dried and sieved through a 2 mm mesh screen. The chemical characterization of the soil samples was done by means of the following analyses: pH in water (1:2.5); P and K extractable by Mehlich 1; exchangeable Ca, Mg, and Al by extraction with 1 mol/L KCl; sum of exchangeable bases (SB); cationic exchange capacity at pH 7.0 (CEC); index of bases saturation (V); index of aluminum saturation (m), and organic matter content (OM).

2.2. Leaching and dissolution/solubilization tests

A composite sample of PG was submitted to leaching and solubilization tests for waste classification. Two waste categories have been established in Brazilian regulations: (1) Class I — dangerous solid waste, with hazardous characteristics such as inflammability, corrosiveness, reactivity, toxicity, and/or pathogeneticity; (2) Class II — harmless solid waste IIA (non-inert) and IIB (inert), wastes that do not present any of the characteristics that were described previously.

2.3. Greenhouse experiments

Experiments to evaluate the use of PG in soil were accomplished in a non-climatized greenhouse with a metallic structure. Soil samples were subjected to lime treatment [10] and were kept humid for 15 d, so as to follow their humidity capacity in the field. They were then mixed with recommended doses of PG, half and twice the recommended amount, in order to verify the effect of this practice on radionuclide bioavailability in both types of soils studied (sandy and clayey).

The PG dosage (recommended mass) equivalent to 1NG (gypsum need) was 0.5 g/dm^3 for clayey soil and 0.2 g/dm^3 for sandy soil, according to criteria established by EMBRAPA [11]. Additional fertilization with phosphorus was done using a dosage of 300 mg/dm^3. For comparison, each experiment was accomplished in triplicate vessels for each type of soil (1NG, 0.5NG, and 2NG PG doses), using a blank vessel (without PG) as a control. After completion of the germination cycle, culture samples were collected, dried at 60°C until constant weight was achieved, ground in a Wiley type grinder, weighed and stored in polyethylene containers for further analyses to be carried out on the edible parts of the plants.

2.4. Analytical methodologies

All analytical methodologies described in this paper were applied according to procedures described in Ref. [12]. UV-Vis spectrophotometry with Arsenazo III was the technique employed to determine uranium and thorium activity concentration in samples of PG and leached water. For uranium (VI), extraction was done with tri-n-butyl-phosphate (TBP), in the presence of $Al(NO_3)_3$ saline complexion agent, EDTA and tartaric acid. Uranium re-extraction in the organic phase was carried out with Arsenazo III solution, forming a red-violet stable complex with absorbance at 650 µm [13].

The procedure for thorium determination was based on separation from impurities by extraction with trioctylphosphine oxide (TOPO), re-extraction with oxalic acid, and colorimetric reaction with Arsenazo III. A stable complex with absorbance read at 665 µm was formed.

Determination of the ^{232}Th activity concentration was carried out by the k0-AAN method for neutron activation [14]. In this case, 200 mg of each sample were weighed in polyethylene tubes, sealed and irradiated in a TRIGA Mark I IPR-R1 reactor located at CDTN/CNEN, using 100 kW with a thermal neutron flow equivalent to 6.35×10^{11} neutrons·cm^{-2}·s^{-1} for 8 h. After irradiation and an appropriate time for decay, the procedure followed medium and long half-life radionuclide determination. Gamma spectrometry was done using an HPGe detector with 15% efficiency. KAYZERO/SOLCO software was used for calculating the concentration of elements.

The ^{238}U activity concentration was determined using the retarded neutron fission activation method, and samples were irradiated with a flow of thermal neutrons. This method uses fast irradiation followed by a reading of retarded neutrons. Samples were irradiated for 50 s, followed by a 30 s allowance for decay time and a 60 s counting time. The irradiation process and the counting utilized an automated pneumatic system. A $^{10}BF_3$ detector was used to count retarded neutrons. The uranium concentration was calculated through linear regression, adjusted according to established patterns.

Alpha spectrometry was used for the determination of U and Th isotope concentrations in plants. Lettuce, corn and soya bean samples were chemically digested by multi-acid attack. The final residue was dissolved with 8M HNO_3 under heating. Thorium isotope separation was done through an anionic DOWEX 1 × 2 resin with concentrated HCl. Uranium isotope separation was done by a UTEVA resin with 0.01M HCl. Both aliquots were dried and electrodeposited for 1 h in previously polished silver planchets, under 1 A and 1.2 A currents, for thorium and uranium, respectively. The quantification of ^{232}Th, ^{229}Th, ^{238}U, and ^{232}U was accomplished in an Alpha Analyst System, Canberra model, with surface barrier semiconductor detectors of 450 mm^2 active area.

The determination of ^{210}Po was also done by alpha spectrometry, according to the procedure described in Ref. [15]. Before dissolution with concentrated mineral acids, a known amount of ^{209}Po was added to the samples in order to obtain the ^{210}Po chemical recovery. The final residue was dissolved in 1.5M HCl and 0.5 g of ascorbic acid was added. Polonium isotopes spontaneously deposit in silver planchets from an acidic medium adjusted to the 80–90ºC range, during a period of 4 h under constant agitation. For leached water samples, radionuclide co-precipitation with iron hydroxide was done to eliminate the main interfering elements. After decanting, the final precipitate was treated as described above.

Gamma spectrometry was employed for ^{226}Ra, ^{228}Ra, and ^{210}Pb measurement in soil and PG. Samples were ground to a No. 80 granulometry. They were then sealed in acrylic containers and analysed by gamma spectrometry after 30 d, enough time to ensure radioactive equilibrium between ^{226}Ra and its progeny ^{214}Pb and ^{214}Bi. An HPGe Canberra detector with a 45% relative efficiency was employed, along with Genie 2000 version 3.1 software for spectral analysis. The photopeaks of 609 and 1020 keV from ^{214}Bi and 351 keV from ^{214}Pb were used to measure ^{226}Ra. A photopeak of 911 keV equivalent to ^{228}Ac (half-life 6.12 h) was used to determine ^{228}Ra. A characteristic photopeak of 46.5 keV was used to measure ^{210}Pb, which also underwent self-absorption corrections.

Radiochemistry was employed for ^{226}Ra, ^{228}Ra, and ^{210}Pb determination in plant and leached water samples. Barium and lead carriers were added. Solid samples suffered a multi-acid chemical digestion, while liquid samples were filtered. The method was based on radium and lead separation from other elements present and separation from each other by selective precipitation, with Ra being co-precipitated as $Ba(Ra)SO_4$ and Pb as $PbCrO_4$. Determinations of Ra and Pb were accomplished through total alpha and beta counting, respectively, in an ultra-low background proportional gas flow counting system (Canberra, Model S5XLB Tennelec).

3. RESULTS AND DISCUSSION

3.1. Phosphogypsum classification and characterization

PG is classified as Class IIA: non-hazardous, non- corrosive and non-inert. Its chemical composition revealed the predominant presence of calcium and sulphur (40.12% CaO and 58.12% SO_4). The percentage ranges for these parameters in phosphate fertilizer are 10–28% and 15–20%, respectively, showing that PG may be employed as a calcium and sulphur source in agricultural activities. Granulometry analysis showed that PG is mainly composed of gypsum ($CaSO_4 \cdot _2H2O$).

3.2. Soil characterization

Soil fertility and granulometry analysis showed that red-yellow latosol was mainly composed of clay, being classified as clayey, while yellow latosol was composed of fine sand and classified as sandy. Both soils were acidic, with low fertility.

3.3. Radionuclide activity concentration in PG and soil samples

The radionuclide activity concentrations in the PG and soil samples are given in Table 1. In general, the PG generated by the phosphate industry in Brazil has radionuclide activity concentrations well below those observed in other countries [3, 4]. The ^{238}U concentration in PG was well below the concentration found in the clayey soil. It is important to mention that:

(a) PG samples usually have low ^{238}U concentrations compared with other radionuclides of natural origin. In phosphate rock, the ^{238}U and ^{232}Th decay series radionuclides are in equilibrium. Following the digestion of

TABLE 1. ACTIVITY CONCENTRATIONS IN PG AND SOIL

	Activity concentration (Bq/g)		
	PG	Sandy soil	Clayey soil
^{238}U	0.080 ± 0.020	<0.050	0.150 ± 0.025
^{232}Th	0.111 ± 0.013	0.036 ± 0.001	0.117 ± 0.003
^{226}Ra	0.252 ± 0.026	<0.020	0.069 ± 0.005
^{228}Ra	0.226 ± 0.029	0.034 ± 0.002	0.114 ± 0.005
^{210}Pb	0.206 ± 0.029	0.020 ± 0.005	0.050 ± 0.008
^{210}Po	0.230 ± 0.026	<0.013	0.043 ± 0.006

phosphate rock by acid, the phosphoric acid produced is enriched with ^{238}U, while ^{232}Th, isotopes of radium and ^{210}Pb tend to concentrate in the PG [2];

(b) Brazilian phosphate rock is of igneous origin, with ^{238}U concentrations lower than those of ^{232}Th;

(c) The clayey soil used in this study was formed from the weathering of granite rocks more than 2700 million years old, which typically exhibit a ^{238}U content of 5–10 ppm.

An average activity concentration of 0.206 ± 0.029 Bq/g was found for ^{210}Pb. A similar value was found for ^{210}Po (0.230 ± 0.026 Bq/g), implying that there was equilibrium between these two radionuclides. Findings from this study confirmed that ^{226}Ra, ^{228}Ra, ^{210}Pb, and ^{210}Po are predominantly incorporated into the PG. For all radionuclides, the activity concentrations in the clayey soil were higher than those in the sandy soil. The average ^{238}U activity concentration in the clayey soil (0.150 ± 0.025 Bq/g) was higher than that in the PG (0.078 ± 0.010 Bq/g). This is due to the fact that this soil comes from a region characterized by sedimentary rocks formed from weathering of granitic rocks and enriched with uranium [16].

According to UNSCEAR [17], the activity concentrations of ^{238}U, ^{228}Ra and ^{226}Ra in soil are in the ranges of 0.016–0.110, 0.011–0.064 and 0.017–0.060 Bq/g, respectively. Evidently the precise value depends on the site geological characteristics. As normally observed in Brazilian soil, the ^{228}Ra activity concentrations in the clayey and sandy soils (0.114 ± 0.005 and 0.034 ± 0.002 Bq/g, respectively) were higher than the corresponding values for ^{226}Ra (0.069 ± 0.005 and <0.020 Bq/g).

In this study, the average activity concentration of ^{210}Pb was found to be 0.050 ± 0.008 Bq/g in clayey soil and 0.020 ± 0.005 Bq/g in sandy soil. The corresponding ^{210}Po activity concentrations were 0.043 ± 0.006 and <0.013 Bq/g. Reference material provided by the IAEA (IAEA–Soil 7) was also analysed in order to provide some form of quality control. The results obtained were within the specified value range for the reference material, confirming that the accuracy of the method was acceptable.

Studies on soils fertilized with PG in Greece and some cultivated products are reported in Ref. [4]. The activity concentrations of ^{226}Ra, ^{228}Ra, and ^{210}Pb in soils were reported to be in the ranges 0.050–0.479, 0.028–0.055 and 0.105–0.585 Bq/g, respectively. Such values are much lower than those found in this study for soils fertilized with PG in Brazil; except for ^{228}Ra, which exhibited similar values to the average found for sandy soil (0.034 Bq/g). The ^{226}Ra activity concentrations in clayey and sandy soils were in the ranges of 0.049–0.058 and <0.020 Bq/g, respectively. The average ^{210}Pb activity concentrations for clayey and sandy soil were 0.047 and <0.030 Bq/g, respectively. The activity concentrations of ^{210}Po in clayey and sandy soil were 0.038–0.042 and <0.011 Bq/g, respectively. For clayey soil, the ^{238}U and ^{232}Th activity concentrations were 0.125–0.142 and 0.109–0.116 Bq/g, respectively. For sandy soil, the activity concentrations of ^{238}U and ^{232}Th were <50 and 0.031 Bq/g, respectively.

3.4. Radionuclide activity concentrations in plants and leached water

The maximum activity concentrations in lettuce found in this study were 0.002 Bq/g for ^{226}Ra, 0.008 Bq/g for ^{228}Ra and <0.008 Bq/g for ^{210}Pb. The maximum value found for soya bean in this study was 0.009 Bq/g. These results indicate that the use of PG may result in radionuclides becoming less concentrated in plants compared with the use of other fertilizers.

Activity concentrations found in leached water samples were very low, most of them below the minimum detectable activity (<0.00011 Bq/g for ^{238}U, <0.000006 Bq/g for ^{232}Th and ^{210}Po, and <0.00002 Bq/g for ^{226}Ra, ^{228}Ra, and ^{210}Pb). There was practically no difference observed between values for the sample with maximum PG dosage and the reference sample (without PG), which indicates that contamination of both soils by PG (sandy and clayey), for all dosages considered, did not result in radionuclide leaching into soil profiles.

3.5. Estimation of soil to plant transfer factors

The transfer factor is the ratio between the radionuclide concentration (dry weight) in the plant and that in the soil, in the root zone. In this study, transfer factors were calculated for those conditions in which the activity concentrations were found to be above the minimum detectable activity. Thus only ^{232}Th and ^{228}Ra transfer factors were obtained for sandy soil.

According to Ref. [18], the fact that transfer factors are larger in sandy soils can be explained due to the lower organic matter content and low cationic exchange capacity, leading to low radionuclide retention and availability to the plant.

The results obtained in this study indicate that in a general way, radionuclide mobility was low in both soils studied. It must be emphasized that the process involving radionuclide mobility in soil is very complex and depends on many factors involving physical, chemical, and biological soil characteristics. Table 2 presents the transfer factor results for lettuce, corn, and soya bean cultivated in clayey and sandy soil.

4. CONCLUSIONS

PG is classified as Class IIA according to the Brazilian Technical Norm ABNT NBR 10004:2004. Chemical analyses revealed a predominance of

TABLE 2. SOIL TO PLANT TRANSFER FACTORS

PG dosage	Transfer factor					
	^{238}U	^{232}Th	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Clayey soil						
0 NG	0.0016	0.0026	0.10	0.020	0.086	0.077
0.5 NG	0.0016	0.0076	0.17	0.051	0.098	0.053
1.0 NG	0.0022	0.0031	0.22	0.084	0.093	0.029
2.0 NG	0.0010	0.0051	0.15	0.043	0.085	0.048
Sandy soil						
0 NG	0.024	0.20	—	—	—	—
0.5 NG	0.020	0.39	—	—	—	—
1.0 NG	0.029	0.64	—	—	—	—
2.0 NG	0.033	0.58	—	—	—	—

calcium and sulphur (40.12% CaO and 58.12% SO_4), compatible with values found in phosphate fertilizers. The average ^{226}Ra activity concentration in PG (0.252 ± 0.026 Bq/g) was below the limit recommended by the US Environmental Protection Agency for agricultural use (0.37 Bq/g). The activity concentrations of ^{228}Ra, ^{210}Pb, and ^{210}Po were within an order of magnitude of that of ^{226}Ra. The activity concentrations of ^{238}U and ^{232}Th were lower, indicating that decay chain equilibrium is broken during the digestion of phosphate rock. Eventually, 80% of the ^{238}U and 70% of the ^{232}Th end up in the phosphoric acid and 80% of the ^{226}Ra, ^{228}Ra, ^{210}Pb, and ^{210}Po end up in the PG.

Both types of soil in the study had elevated acidity and a low organic matter content; typical of the Cerrado region. Transfer factors obtained for the plants studied varied from 0.00094 to 0.34, considering all radionuclides. Generally, results from this study indicated that the radionuclide mobility in soils was low.

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RADIOLOGICAL ASSESSMENT OF USING PHOSPHOGYPSUM AS BUILDING MATERIAL

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Abstract

In order to assess the feasibility of using phosphogypsum as a building material, an experimental house was built with phosphogypsum panels of different origins. The aim of this study is to assess the external and internal exposure of residents. Phosphogypsum samples were analysed by high resolution gamma spectrometry for their ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K content. The activity concentrations were 0.016–0.393 Bq/g for ^{226}Ra, 0.026–0.253 Bq/g for ^{232}Th and 0.027–0.852 Bq/g for ^{210}Pb. The results for ^{40}K were lower than 0.081 Bq/g. The effective dose was evaluated for each type of phosphogypsum panel. The effective doses due to external exposure were always below 1 mSv/a, the dose limit for the general public. Radon measurements in the house were carried out using the passive method with solid state nuclear track detectors (CR-39) over a period of 15 months. The detectors were changed every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The radon concentration varied from 45 to 119 Bq/m^3. These results are below 200 Bq/m^3, the recommended investigation level for radon in dwellings.

1. INTRODUCTION

It is well known that natural radioactivity in building materials and radiation from the ground constitute the most important sources of radiation exposure [1]. Concerns about health hazards and environmental pollution have heightened the interest in radon levels in building materials. Building materials that may be of radiological significance include marl, blast furnace slag, flyash, phosphogypsum, Portland cement clinker, anhydrite, clay, and granites rich in radium or thorium [2].

Chronic exposure of humans to low levels of radiation can cause health effects which may appear several years after the exposure. The main contributors to external exposure from gamma radiation are the radionuclides of the ^{238}U and ^{232}Th series together with ^{40}K that are present in small quantities in the earth's crust and in building materials. The most important contributors to the internal exposure of the population to natural sources are the short lived decay products of radon (^{222}Rn) and thoron (^{220}Rn).

In recent years, various surveys of the radionuclide content of building materials were published [3–6], in order to estimate the indoor radiation exposure of the occupants. Indoor exposure to gamma radiation and radon inhalation can be enhanced if industrial by-products such as phosphogypsum are used to replace some of the natural components of building materials.

Phosphogypsum is a by-product obtained from the wet acid processing of phosphate rock to produce phosphoric acid. For every tonne of phosphoric acid produced in the reaction of phosphate rock with sulphuric acid, about 4–5 t of phosphogypsum are produced. The Brazilian annual production of phosphogypsum is up to 5.4 million t [7]. All the countries that produce phosphate fertilizer by wet acid processing of phosphate rock are facing the same problem of finding solutions for the safe application of phosphogypsum, in order to minimize the impact caused by the disposal of large amounts of this material. Phosphogypsum can be classified as NORM, and its safe application should comply with specific regulations.

At the international and regional levels, the IAEA and the European Commission (EC), respectively, have recently published recommendations on the application of the concepts of exclusion, exemption and clearance, the scope of application of which includes NORM activities [8–10]. In IAEA Safety Reports Series No. 44 [9], the concepts of exemption and clearance have been applied to bulk amounts of material, by taking into account exposure scenarios that included those described above. These scenarios were applied only to artificial radionuclides, however. For radionuclides of natural origin, the recommended criteria for regulation, 1 Bq/g for ^{238}U and ^{232}Th series radionuclides and 10 Bq/g for ^{40}K, were based on the global distribution of radionuclide activity concentrations in rocks and soil. The rounded exemption and clearance levels established by the EC [11] are 0.5 Bq/g for ^{226}Ra and ^{228}Th; 1 Bq/g for ^{228}Ra; 5 Bq/g for U_{nat}, ^{210}Pb, ^{210}Po and ^{232}Th; and 10 Bq/g for ^{230}Th.[1]

The activity concentrations in Brazilian phosphogypsum for all sources of production [7] are below the criteria for regulation recommended by the IAEA and the exemption levels recommended by the EC. In Brazil, the regulatory agency (Comissão Nacional de Energia Nuclear (CNEN)) has recently published a radiation protection guideline on the mining and processing of NORM that may generate enhanced concentrations of radionuclides [12], according to which the phosphate industry activity is classified as category III on account of the activity concentration in phosphogypsum. In terms of this guideline, the facility should

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[1] The values for ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th include the progeny of these radionuclides.

evaluate the environmental impact of the disposal of phosphogypsum. However, there is not yet a Brazilian guideline giving criteria for exemption and clearance that apply specifically to the use of phosphogypsum. The use of phosphogypsum is very important from the social and economic point of view and also regarding environmental protection. Phosphogypsum can be used as a base for roads, as a building material and in agriculture as a soil amendment.

Only a small proportion of the phosphogypsum produced worldwide (14%) is used as a building material. However, phosphogypsum often contains significant amounts of radioactivity originating mainly from the ^{238}U and ^{232}Th decay series, which, according to Ref. [13], can create a health hazard. Radium, which decays to ^{222}Rn through alpha particle emission, is one the most important radionuclides from the point of view of radiation protection. Exposure to radium and radon originating from phosphogypsum is an important health concern associated with the use or disposal of this material. The main health concern associated with ^{222}Rn arisies from its short lived alpha emitting progeny, which can cause damage to the lungs after chronic exposure. Radon is an inert, noble gas and may become airborne by diffusing into the air. One of the most important sources of indoor radon is the underlying soil; however the contribution from building materials should also be taken into account.

In order to assess the feasibility of using phosphogypsum as a building material, an experimental house was built with phosphogypsum panels (manufactured with phosphogypsum of various origins) in São Carlos, São Paulo State, Brazil. The panels were developed with a new process (patent applied for), denominated UCOS (humidification, compaction and drying), which works with plasters made from both mineral gypsum and phosphogypsum [14]. The plaster prepared through this process may be used inside and outside the building and is sturdy enough to be used in the construction of buildings up to eight storeys high. The house walls are light, easily mounted and have thermal and acoustic insulation. Other advantages of these panels compared to the conventional process are the short construction time (a seven room house such as the experimental house takes only one month to build) and cost, which is approximately 30% lower than that for conventional construction methods.

In the experimental house, two bedrooms and one bathroom were lined entirely with phosphogypsum and designed so as to enable a comprehensive radiological assessment to be performed, including the modelling of the indoor external dose rate and measurement of the external gamma exposure and radon concentration. The walls consisted of double sets of joined panels. The internal cavities between wall panels, as well as between the roof and ceiling, were lined with blanketing material. The floor areas were 9.07 m^2 for the first bedroom, 11.08 m^2 for the second bedroom and 2.4 m^2 for the bathroom. The phosphogypsum panel thickness was 1.5 cm for the walls and 1 cm for the ceiling. The wall cavities were 15 cm wide.

The aim of the study was to assess indoor external exposure and radon concentrations in a house constructed with phosphogypsum panels of various origins. Samples of the material were analysed by high resolution gamma spectrometry for their ^{226}Ra, ^{210}Pb, ^{232}Th, and ^{40}K content. The radium equivalent activity and effective dose from external exposure were also calculated. Radon measurements were carried out using passive solid state nuclear track detectors (CR-39) over a period of 15 months. The detectors were changed every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The results obtained from this study can contribute to the development of national standards and guidelines concerning the safe use and management of this material as a building material.

2. MATERIALS AND METHODS

An experimental house was constructed, in which the walls and ceilings were built of phosphogypsum panels. These panels were manufactured with phosphogypsum from different producers, namely Ultrafertil, located in Cubatão (CT), Bunge, in Cajati (CA) and Fosfertil, in Uberaba (UB). The bathroom and one bedroom were built with phosphogypsum panels from Cubatão and the other bedroom was built with Cajati phosphogypsum panels.

Twelve samples of phosphogypsum panels (five from Cubatão, four from Cajati and three from Uberaba) were crushed and packed in a 100 mL polyethylene flask and sealed for about four weeks prior to the measurement, to ensure that equilibrium was reached between ^{226}Ra and its short lived progeny. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples of phosphogypsum panels were measured by gamma spectrometry with a Eurisys EGNC 15-190-R hyperpure germanium detector. The relative efficiency of the detector was 15% and the effective energy resolution for the 1.33 MeV ^{60}Co gamma transition was 1.9 keV. The detection efficiency curve was calculated for aqueous solutions containing certified activity concentrations of gamma emitters covering a wide range of energies, encompassing the radionuclide energies determined in the samples. Background measurements were taken and subtracted in order to get net counts for the sample. Samples were measured during a period of 40 000–200 000 s, depending on the radioactivity levels in the samples. All spectra were analysed with Eurisys Interwinner 4.1 software for personal computer analysis of gamma spectra from HPGe detectors [15].

The activity concentration of ^{40}K was determined directly by its own gamma peak at 1460.8 keV. The activity concentration of ^{226}Ra was determined using the 295.2 and 351.9 keV gamma emissions from ^{214}Pb and the 609.3 and 1120.3 keV gamma emissions from ^{214}Bi. The activity concentration of ^{232}Th was determined using the 338.5, 911.1 and 968.9 keV gamma emissions from ^{228}Ac and the 727.2 and 1620.6 keV gamma emissions from ^{212}Bi. In a previous paper [7], it was shown that the measurements of ^{232}Th in phosphogypsum samples, analysed by alpha and gamma spectrometry, gave results which are representative of the radionuclide activity. In this paper, the ^{232}Th activity concentration was performed by gamma spectrometry on its progeny gamma emitters. The activity concentration of ^{210}Pb was determined by its 46.5 keV photopeak. A self absorption correction was applied owing to the low energy gamma ray attenuation within the sample. The approach used was the same as that described in Ref. [7]. The minimum detectable activity concentrations in the phosphogypsum panel samples were, for a counting time of 200 000 s, 0.0028 Bq/g for ^{226}Ra, 0.0051 Bq/g for ^{232}Th, 0.039 Bq/g for ^{40}K and 0.018 Bq/g for ^{210}Pb.

Various well established techniques are available for measuring the concentrations of radon and its progeny. The passive method with solid state nuclear track detectors (SSNTDs) has been used widely, since it is regarded as being inexpensive, reliable and unaffected by widely varying climatic conditions [16]. In this study, the ^{222}Rn concentration was obtained by the passive detection method with SSNTDs placed in diffusion chambers. CR-39 in 2.5 cm squares was used as the detection medium. As a diffusion chamber, a hemispherical ‘closed-can’ of 4 cm radius was selected. This chamber consists of a polypropylene holder made of an upper and lower half which snaps together during assembly. The fit of the two halves is quite tight, in order to exclude moisture, dust and radon progeny. Thoron is excluded from the diffusion chamber because of its short half-life and diffusion time. The detectors were suspended in the dwellings from the ceiling at a height of 1.7 m from the ground and placed away from any surface to avoid any plate-out effects. After exposure for approximately 3 months, the detectors were collected and replaced by fresh ones. The detectors were etched in a KOH solution (30% mass concentration) at 80ºC for 5.5 h in a constant temperature bath. After etching, the detectors were washed, dried and scanned under a microscope for track density measurements. The background was 220 ± 16 tracks/cm^2. Using a calibration factor of 0.0216 ± 0.0015 tracks/cm^2 per Bq·d·m^{-3}, obtained with a Pylon model RN-150 calibrated radon gas source; the track density was converted to radon concentration in the environment.

The radon concentration was calculated from the following equation [17]:

$$C = \frac{D}{k \cdot t} \tag{1}$$

where C is the radon concentration (Bq/m^3), D is the track density (tracks/cm^2), k is the calibration factor (tracks/cm^2 per Bq·d·m^{-3}) and t is the exposure time (d). Radon measurements were carried out over a period of 15 months, changing the detectors every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The detectors were placed in the two bedrooms and the bathroom.

3. RESULTS AND DISCUSSION

3.1. Activity concentration and radium equivalent activity

Phosphogypsum panels from three production locations, Cubatão (CT), Cajati (CA) and Uberaba (UB) were analysed for ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K activity concentrations. The radium equivalent activity (Ra_{eq}) is a common index that compares the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in building materials, taking into account the radiation hazards associated with them. This index is based on the assumption that 1 Bq/g of ^{226}Ra, 0.7 Bq/q of ^{232}Th and 13 Bq/g of ^{40}K produce the same gamma dose rate. According to UNSCEAR recommendations [1, 18], the maximum value of Ra_{eq} in building materials must be less than 0.37 Bq/g in order to keep the dose from external gamma exposure below 1.5 mSv/a. The radium equivalent activity provides a useful guideline on radiation protection for the general public in dwellings. It was calculated as follows:

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.007\ C_K \tag{2}$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively (Bq/g). Table 1 presents the average activity concentrations and standard deviation for ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K in the phosphogypsum panels used in the house construction. The radium equivalent activity is also presented.

The activity concentrations varied widely for the three phosphogypsum origins. Panels from Cubatão (CT) showed the highest activity concentrations for all the radionuclides analysed, while the lowest values were observed in phosphogypsum from Cajati (CA). The activity concentrations in the products

TABLE 1. ACTIVITY CONCENTRATIONS IN PHOSPHOGYPSUM PANELS

Origin of panel	Average activity concentration (Bq/g)				
	^{226}Ra	^{232}Th	^{40}K	^{210}Pb	Radium equivalent
CA	0.016 ± 0.001	0.026 ± 0.003	<0.039	0.026 ± 0.007	0.056
CT	0.392 ± 0.10	0.253 ± 0.003	<0.081	0.852 ± 0.138	0.759
UB	0.294 ± 0.003	0.151 ± 0.006	<0.056	0.295 ± 0.016	0.513

from Cajati are lower than the worldwide average values for building materials of 0.05 Bq/g for ^{226}Ra, 0.05 Bq/g for ^{232}Th and 0.5 Bq/g for ^{40}K [18]. All the phosphogypsum panels exhibited ^{40}K concentrations lower than the world averages for building materials. The results obtained in this work are in agreement with national and international values published in the literature for phosphogypsum [7, 19].

There is a great variation in the radium equivalent activity results, depending on the origin of the phosphogypsum. The highest values were reported for Cubatão (CT) and Uberaba (UB) panels, both of which exceeded the value recommended by UNSCEAR (0.37 Bq/g), while the panels from Cajati (CA) are lower than this value. However, it should be noted that the radium equivalent activities obtained from the maximum activity concentrations in industrial by-products used for building materials in the European Union reaches 1.351 Bq/g [10], a figure significantly higher than the maximum value of 0.759 Bq/g obtained in the present work for Brazilian phosphogypsum.

3.2. Effective dose for external exposure

The effective dose received indoors from external exposure to phosphogypsum panels was assessed according to UNSCEAR procedures through the standard room concept, according to the equation:

$$E = T\, f\, b \times 10^{-6}\, (q_{Ra}C_{Ra} + q_{Th}C_{Th} + q_K C_K)\, m \tag{3}$$

where E is the effective dose from external exposure; T is the exposure time (8760 h/a); f is the fraction of time spent indoors (0.8); b is the conversion factor from absorbed dose in air to effective dose (0.7 Sv/Gy); q_{Ra}, q_{Th} and q_K are the conversion factors from ^{226}Ra, ^{232}Th and ^{40}K activity concentrations to absorbed dose in indoor air, respectively (nGy/h per Bq/g); C_{Ra}, C_{Th} and C_K are the ^{226}Ra,

^{232}Th and ^{40}K activity concentrations, respectively (Bq/g) and m is the mass fraction of material in the standard room. The choice of conversion factor from activity concentration to absorbed dose is the most important factor for evaluating the external dose from building materials. Gamma radiation from walls is strongly dependent on the wall thickness and material density, so it is useful to adopt the standard room concept to estimate the dose. Values of q_i reported in Ref. [20] were used as the basis for applying a previously developed computational model [21] to forecast external doses indoors using the parameters of the experimental house. The q_i values take into account the gamma transitions of ^{40}K and radionuclides in the ^{226}Ra and ^{232}Th decay series, the wall thickness of the house and the density of the phosphogypsum panels. The estimated effective dose from external exposure for each source of phosphogypsum panel, is presented in Table 2. For all origins, the effective dose from external exposure was lower than the 1 mSv annual dose limit for members of the public.

3.3. Radon concentration

The ^{222}Rn concentrations in the experimental house were calculated from the SSNTD measurements using Eq. (1). The average radon concentrations and standard deviations obtained for the period May 2007 to September 2008 are shown in Table 3. The radon concentrations varied from 45 to 119 Bq/m^3 in the bedrooms and from 83 to 105 Bq/m^3 in the bathroom. The difference in radon concentration between bedrooms 1 and 2 can be explained by the origin of the phosphogypsum. Bedroom 1 and the bathroom were built with phosphogypsum from Cubatão (average ^{226}Ra concentration 0.392 ± 0.010 Bq/g) and bedroom 2 was built with phosphogypsum from Cajati (average ^{226}Ra concentration 0.016 ± 0.001 Bq/g). Reference to Fig. 1 shows that there was no clear seasonal variation of indoor radon concentration.

TABLE 2. EFFECTIVE DOSE FOR EXTERNAL EXPOSURE IN A STANDARD ROOM

Origin of panel	Annual effective dose (mSv)
CA	0.02
CT	0.20
UB	0.14

TABLE 3. RADON CONCENTRATIONS IN THE EXPERIMENTAL HOUSE

	^{222}Rn activity concentration in air (Bq/m^3)				
	May to August 2007	August to November 2007	November 2007 to February 2008	February to June 2008	June to September 2008
Bedroom 1	98 ± 14	105 ± 11	99 ± 5	119 ± 9	106 ± 11
Bedroom 2	—	46 ± 2	50 ± 2	45 ± 2	48 ± 3
Bathroom	88 ± 13	105 ± 8	99 ± 7	83 ± 8	86 ± 9

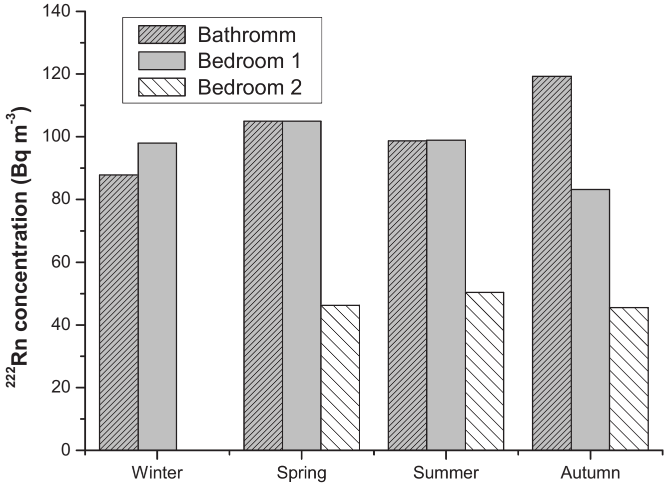

FIG. 1. Seasonal variation of indoor ^{222}Rn concentration in the experimental house.

There was a good correlation between the indoor ^{222}Rn concentration and the ^{226}Ra concentration in the phosphogypsum panels (see Fig. 2). Therefore, the ^{222}Rn concentration in a room built with panels from another source can be predicted from the ^{226}Ra concentration in those panels and the slope of regression line in Fig. 2.

4. CONCLUSIONS

The activity concentrations of ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K in phosphogypsum panels from Cubatão, Cajati and Uberaba have been measured by gamma spectrometry. The radium equivalent activity and the effective dose for

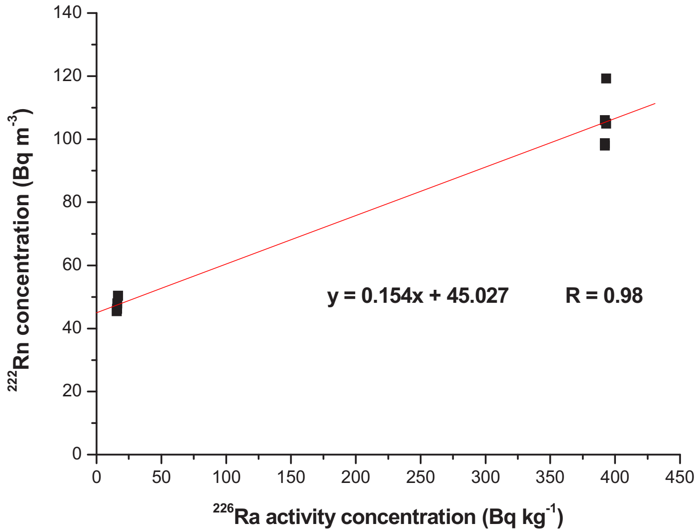

FIG. 2. Relationship between indoor radon concentration and ^{226}Ra activity concentration in the phosphogypsum panels.

external exposure were assessed from the activity concentrations. The results showed that the phosphogypsum panels from Cubatão exhibited the highest radioactivity levels, while those from Cajati exhibited very low activity concentrations and, therefore, negligible risk.

There is no regulatory framework for the use of NORM as a building material in Brazil. Nevertheless, it is possible to adopt 1 mSv/a as a reference level for protection of the public against radiological impact of naturally occurring radioactive materials, as recommended by the International Commission on Radiological Protection (ICRP) [22]. According to EC guidance [10], the annual effective dose due to gamma radiation in dwellings should not exceed 0.3 mSv for the building materials to be exempted from all restrictions concerning their radioactivity. All phosphogypsum panels evaluated in this study gave rise to effective doses for external exposure lower than this recommended value. The present work showed that, according to dose criteria recommended by the ICRP and the EC, the phosphogypsum panels do not pose any significant additional health risk to dwellers and its use can be considered to be safe for inhabitants.

The ICRP recommends that the action level for radon in dwellings should be in the range 200–600 Bq/m^3 [23]. The radon concentrations determined from this study are below 200 Bq/m^3. It should be noted that the radon concentration results took into account the radon from soil under the building.

The results obtained in this study point to the fact that the radon concentrations in dwellings made from phosphogypsum panels are comparable with those in houses built from conventional materials [1, 24]. Therefore, the use of phosphogypsum as a building material poses no additional health risk to occupants.

ACKNOWLEDGEMENTS

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PHYSICO-CHEMICAL AND RADIOACTIVE CHARACTERIZATION OF CO-PRODUCTS FROM THE TITANIUM DIOXIDE NORM INDUSTRY FOR THEIR USE IN CEMENT MANUFACTURING

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Abstract

This paper presents the results of a study to characterize the raw materials (ilmenite and slag), residue (red gypsum) and co-products (ferrous sulphate monohydrate and heptahydrate) associated with titanium dioxide production, in particular their elemental composition (major, minor and trace elements), mineralogy and radioactive content, with the objective of applying this knowledge to facilitate the use of some of these materials in applications such as construction and civil engineering. Obviously, the environmental and health impact of the co-products should comply with existing regulations. In particular, the main properties of cement produced with different proportions of red gypsum and the improvements obtained in relation to ordinary Portland cement have been studied. It is demonstrated that the levels of pollutants in the red gypsum cement remain within safety limits.

1. INTRODUCTION

The use of residues generated in the majority of industrial production processes (rather than their disposal as waste) is the subject nowadays of more and more research because, although the protection of health and the environment is of great importance, the economic benefits accruing from residue use cannot be neglected either [1, 2]. The minimization of the disposal of industrial residue as waste avoids its direct release to the environment, generating health and environmental benefits in several industrial processes.

Moreover, in addition to the generation of the main product, the appropriate treatment of a fraction of the residue generated could lead to the production of co-products with economic value and broad applications [3]. One well known use of industrial residues is through their incorporation into cement [4–7]. The use of such residues in the cement industry is performed in two ways: the residue is either used as a component of the clinker, which then forms the basis for the cement, or is added subsequently to the clinker in the cement production process.

This paper concerns one particular NORM industry that endeavours to make wide use of its residues. It is located 12 km from the city of Huelva in south-western Spain and produces titanium dioxide pigments and two co-products obtained as a consequence of the treatment of residues generated by the process.

Two raw materials are used as feedstock in the titanium dioxide production at the Huelva factory: ilmenite ($FeTiO_3$) and slag. Ilmenite is a heavy mineral containing approximately 43–65% titanium dioxide [8], and can be considered as NORM because it generally contains enhanced amounts of uranium and thorium as a result of its geological origin. The titaniferous slag, which contains 70–80% titanium dioxide, is a co-product resulting from the smelting of ilmenite [9]. The oldest and most common process for titanium dioxide production is the sulphate process (see Fig 1). The main process steps are as follows:

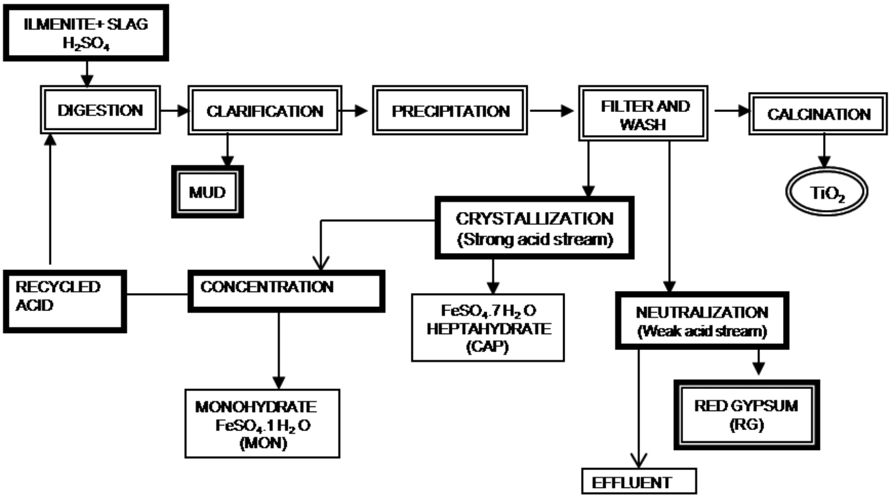

FIG. 1. The sulphate process used for TiO_2 production in the Huelva plant.

(i) *Digestion of the ore (batch operation)*: A carefully controlled blend of ilmenite and slag is mixed with highly concentrated sulphuric acid (80–95%) to digest the TiO_2-containing feedstock. The resulting liquor contains titanyl sulphate ($TiOSO_4$) and iron sulphate ($FeSO_4$) dissolved in sulphuric acid. To ensure that all the Fe is in a dissolved state, the liquor is passed through a bath of scrap metal (Fe reduction step).

(ii) *Clarification of the resulting liquor*: The reduced liquor flows into a clarification tank where the undissolved solids (mud) are separated from the solution by flocculation and filtration.

(iii) *Titanium dioxide precipitation*: The clarified liquor is hydrolyzed to precipitate hydrated titanium.

(iv) *Hydrated TiO_2 separation*: The hydrated TiO_2 after its precipitation is separated from the mother liquor by vacuum filters (Moore filters).This liquor can be considered as an intermediate product and is treated for the generation of two co-products, as described later in the paper.

(v) *TiO_2 washing*: After the separation of the mother liquor, the filtered TiO_2 cake is washed with water to remove the remaining impurities. The generated weak acid solution used in this final wash can, in principle, be considered as a residue from the process.

(vi) *Preparation of TiO_2 product*: The TiO_2 pulp is placed in rotary kilns for the removal of its water content and some traces of sulphur. The resulting solid is cooled, milled, coated, washed, dried and finely ground (‘micronized’) before being packed for commercial distribution.

Initially, the mother liquor (‘strong’ acid, 20–25 % H_2SO_4) is pumped into batch cooler crystallizers, where the iron is removed as solid ferrous sulphate heptahydrate ($FeSO_4 \cdot 7H_2O$). This constitutes the first co-product of the TiO_2 process, commonly known as copperas (CAP). The remaining strong acid is then reconcentrated for recycling back to the initial digestion step. During this process, ferrous sulphate monohydrate (MON) is precipitated, forming a second co-product that is separated by filtration. Then, the resulting clean sulphuric acid can be recycled back to the ilmenite digestion step.

The weak sulphuric acid coming from the washing of the TiO_2 pulp is also processed by sending it to a neutralization plant where, by the addition of lime or limestone, a solid residue called red gypsum (RG) is generated. RG comprises mainly calcium sulphate di-hydrate ($CaSO_4 \cdot 2H_2O$) and iron hydroxides, which give it its red colour.

Annually, about 142 000 t of raw material are processed at the Huelva plant (85% ilmenite and 15% slag), leading to the generation of 70 000 t of RG, 140 000 t of CAP and 125 000 t of MON.

Until now, the RG has not been put to any use and has been disposed of in an authorized area. This approach is costly, so the company decided to re-evaluate its options. Large amounts of natural gypsum are used as a setting retardant in cement production, by adding it to the clinker in the proportion 3–5% [10]. The possibility of using red gypsum as a substitute for natural gypsum in this application was evaluated.

2. MATERIALS AND METHODS

Samples of raw materials (ilmenite and slag), residue (red gypsum) and co-products (ferrous sulphate heptahydrate and ferrous sulphate monohydrate) were collected from the plant. Five sampling campaigns were organized during a period of 1 month, in order to analyse the possible time variability in the industrial process. After collection, the raw materials were dried at 105°C until reaching a constant weight, while the co-products and RG were dried at 45°C to avoid the loss of their hydration water.

As the objective of the work was to check the possibility of substituting red gypsum for natural gypsum as a setting retardant in the manufacture of cement, red gypsum was added to conventional clinker in three proportions: 2.5% (YA), 5% (YB) and 10% (YC). The properties of these mixtures were compared with those of a commercial cement (CEM), which falls within the 52.5 N/mm^2 resistance category and comprises a mixture of 97% clinker and 3% natural gypsum.

The mineralogical compositions were analysed in the raw materials, co-products and residue by means of the X ray diffraction (XRD) technique. The concentrations of major elements, heavy metals, and other trace elements were determined by X ray fluorescence (XRF) and ICP–MS. Additionally, the activity concentrations of radionuclides of natural origin in these materials were determined by both alpha and gamma spectrometry with semi-conductor detectors. Also, granulometric analyses were performed, because the grain size is very important for the manufacture of cement.

3. RESULTS AND DISCUSSION

3.1. Physicochemical characterization of the materials

The radionuclide activity concentrations are given in Table 1. The combined activity concentration of ^{238}U and ^{232}Th in ilmenite is about 0.5 Bq/g. The values for slag are lower than those found in typical undisturbed soil

TABLE 1. ACTIVITY CONCENTRATIONS IN PROCESS MATERIALS

	Water content (%)	Activity concentration (Bq/g)				
		^{238}U	^{226}Ra	^{232}Th	^{228}Ra	^{40}K
Ilmenite	4	0.095 ± 0.010	0.110 ± 0.010	0.420 ± 0.015	0.440 ± 0.030	0.030 ± 0.005
Slag	3	0.0059 ± 0.0006	0.0061 ± 0.0006	0.014 ± 0.001	0.0090 ± 0.0004	nd
CAP	40.3	0.0015 ± 0.0002	nd[a]	0.013 ± 0.002	0.004 ± 0.001	nd
MON	4.2	0.053 ± 0.002	0.0091 ± 0.0004	0.365 ± 0.013	0.043 ± 0.002	nd
RG	46.3	0.020 ± 0.001	0.014 ± 0.001	0.127 ± 0.003	0.091 ± 0.003	0.012 ± 0.002

[a] nd = not detected.

(0.02–0.03 Bq/g [11]). The CAP activity concentrations are less than 0.01 Bq/g, so the use of this material is not restricted by its radioactivity content. MON has higher ^{232}Th concentrations, as well as an appreciable fraction of the initial U that enters the process with the ilmenite. The radioactivity content of RG is moderate, indicating that a minority fraction of the content in the treated raw material is accumulated in this potential co-product.

Ilmenite contains mainly Fe_2O_3 (44 %) and TiO_2 (~50 %), with low percentages of SiO_2 (0.7 %), MnO (1.3 %) and MgO (0.33 %) [8]. Slag has a higher titanium content (75 % TiO_2) and a lower iron content (~11 % F_2O_3) [12]. CAP ($FeSO_4{\cdot}7H_2O$) and MON ($FeSO_4{\cdot}1H_2O$) contain high percentages of iron (39 and 31 %) and sulphur (22 and 25%), as corroborated by XRD analyses. As a result of its formation process, CAP (formed by crystallization) contains a lower proportion of impurities than MON (formed by precipitation).

The levels of radionuclides and heavy metals in CAP and MON are not a problem in present commercial applications. Copperas (CAP) is currently being used as a basic soil amendment, animal feed and a primary flocculant for waste water treatments. As for the monohydrate (MON), it is valued as a fertilizer for soils that are poor in iron.

RG contains 27% SO_3 and 33% CaO, with a significant iron hydroxide content (12%) giving it its characteristic dark red colour [13, 14]. This co-product has a significant titanium content (~7 % TiO_2), which has led the industry to seriously investigate ways to recover it. The diffractogram obtained for RG indicated, as expected, that the main crystalline phase is $CaSO_4{\cdot}2H_2O$. Moreover, this diffractogram is very similar to that found in the literature for natural gypsum or phosphogypsum [15]. In addition, the granulometric data obtained shows that the particle size distribution for RG reaches a maximum at around 40 μm. This is slighter higher than the corresponding value for commercial cement (about 30 μm) and is twice that of clinker (about 20 μm), giving greater confidence in the use of RG as a substitute for natural gypsum in cement. Research is currently focused, therefore, on replacing natural gypsum in cement with red gypsum as a setting retardant.

3.2. Tests to assess the viability of cements with red gypsum additive

The first step after the formation of the three dried mixtures of clinker with red gypsum, YA, YB and YC (see Section 2) was to determine the appropriate water/cement (W/C) ratio in weight terms for obtaining a ‘normal’ consistency of the paste formed, as defined in the UNE-EN 196-3 regulation in Spanish law. A normalized Vicat apparatus was used for this purpose. All the W/C ratios are quite similar (see Table 2), with no significant differences found between the commercial and red gypsum cements.

TABLE 2. COMPARISON OF CEMENTS MADE USING RED GYPSUM WITH COMMERCIAL CEMENT

	W/C	Bending strength (MPa)		Compressive strength (MPa)		Setting time (min)	
		2 d	28 d	2 d	28 d	Initial	Final
CEM	0.27	6.8 ± 0.3	10.1 ± 1.2	34.4 ± 0.4	61.3 ± 1.0	139	224
YA (2.5% RG)	0.29	4.2 ± 0.15	9.8 ± 0.6	17.8 ± 0.3	51.7 ± 0.6	82	129
YB (5% RG)	0.27	5.4 ± 0.25	8.2 ± 2.7	23.1 ± 0.3	57.9 ± 1.0	108	298
YC (10% RG)	0.29	7.6 ± 0.8	10.8 ± 0.8	31.5 ± 0.8	59.6 ± 1.5	216	351

Furthermore, in order to analyse the possible risks of short-time expansion, which can affect the red gypsum cements formed, the Le Chatelier test was performed following the UNE-EN 196-3 regulation. The results showed that the presence of red gypsum does not cause any significant modification in the expansion behaviour of the cement in comparison with that of commercial cement, irrespective of the proportion of red gypsum used as an additive. More importantly, the values comply with Spanish legislation.

For the mechanical test, mortars (sand and cement mixed in a ratio of 3:1) were used to conduct tests to evaluate the compressive and flexural strength of the cements formed with red gypsum. The resistances to flexing and compression determined through these tests for the three red gypsum cements assessed in this work are given in Table 2, which shows also the comparative results for the commercial cement (CEM).

The setting times for the three red gypsum cements are also comparable to those obtained for the commercial cement (see Table 2). The data show that the addition of red gypsum in the proportions used in this work leads to setting times that are within those required by national legislation, and it is noteworthy that the initial and final setting times are prolonged by increasing the proportion of red gypsum used as an additive in cement fabrication, further underlining the role of red gypsum as a retardant in the hardening of cement. Adding higher percentages of red gypsum extends the initial and final setting times.

The mechanical behaviour of the RG cements improves with increasing RG content, thereby approaching the performance of the commercial cement. The mechanical performance of the YC sample (10% red gypsum) is similar to that of the commercial cement. This is important, as it supports the use of RG as an additive in the formation of cements because it is possible to reduce the amount of clinker used, with all the economic benefits (cost savings) that this entails. In this regard, it should be remembered that the commercial cement taken as a reference in the study contains approximately 97% clinker and 3% natural gypsum. In addition, it is important to note that, in the mineralogical characterizations for the three RG mixtures performed 28 days after production, the results are similar to those obtained for the comercial cement (CEM).

Finally, the environmental impact of the use of red gypsum in cement manufacturing has been assessed. Table 3 shows the concentrations of various metallic elements in RG compared with those in soil and with the critical concentration values that define the ecotoxicity threshold (meaning that if the concentration of an element is lower than this threshold, it does not generate a significant environmental impact [16]). Except possibly for Cr, the metal concentrations in red gypsum are lower than the ecotoxicity threshold. Given that the maximum amount of red gypsum in cement would be about 10%, even the Cr concentration in the cement will be well below this threshold. It can be concluded therefore, that the use of red gypsum as an additive in cement does not pose a threat to the environment.

The European Commission recommends that an activity concentration index be used as an investigation level for screening of building material that might be of concern from a radiological point of view, pending a separate dose assessment specific to the scenario in which the material is used [18]. The activity concentration index (I) is derived as follows:

$$I = \frac{C_{Ra}}{0.3} + \frac{C_{Th}}{0.2} + \frac{C_K}{3}$$

TABLE 3. COMPARISON OF METALLIC ELEMENT CONCENTRATIONS

	RG (ppm)	Soil, continental crust (ppm) [17]	Critical concentration (ppm)
As	10	4.8	20–50
Cd	1	0.09	3–8
Co	16	17	25– 50
Cr	109	92	75–100
Cu	8	28	60–125
Ni	30	47	100
Pb	19	17	100–400
Zn	212	67	70–400

where C_{Ra}, C_{Th} and C_K are the radium, thorium and potassium activity concentrations (Bq/g) in the building material. For materials used in bulk amounts, it is recommended that the activity index should not exceed a value of 1 in order to limit the dose to 1 mSv/a. In the case of red gypsum with the activity concentrations given in Table 1, it can be seen that, when applying this radiological criterion, red gypsum can be used as a component of construction materials in any proportion with no radiological consequences.

4. CONCLUSIONS

The present study has been carried out to acquire detailed information on the composition of raw materials, co-products and residue from the titanium dioxide pigment production process. The physical, chemical and radiological characterization of these materials has confirmed that titanium dioxide production is a NORM industry and that the concentrations of metals and radionuclides in the CAP and MON co-products (ferrous sulphate heptahydrate and monohydrate, respectively) are within the European regulations in relation to their applications.

Furthermore, it is concluded that red gypsum can be safely applied as a substitute for natural gypsum in the fabrication of commercial cements without decreasing the quality of the cements generated and without causing any environmental or radiological impact.

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RADIATION EXPOSURE FROM THE USE OF NORM IN BUILDING MATERIALS IN GERMANY

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Abstract

The use of NORM in building materials has been under regulatory control in Germany since 2001. Based on an incremental effective dose of 1 mSv per year, the allowable concentrations of radionuclides in residues from specific industrial processes and their content in building materials are defined. At the European Union (EU) level, regulations on natural radioactivity in building materials will be introduced within the framework of the current revision of the European Council Directive 96/29/Euratom. The regulations will be based on the recommendations contained in the European Commission publication Radiation Protection 112 and will not be restricted only to the use of NORM. The dose criterion in Radiation Protection 112 of 1 mSv per year refers to external gamma exposure only. The current NORM regulations in Germany explicitly consider the radon inhalation pathway also. Also, the Federal Office for Radiation Protection (BfS) is recommending a target level of 100 Bq/m³ for radon concentration in homes. In view of the coming legal changes at the EU level concerning building materials, an up to date overview of the current exposure situation, including the incorporation of NORM into building materials, appeared to be necessary. Between 2007 and 2009**,** the BfS, in collaboration with the German Building Materials Association, investigated natural radioactivity in different types of current building materials in Germany and the resulting doses, including the contribution from radon inhalation. For classical building materials such as brick and lightweight concrete, exposures may be as high as 1 mSv/a via external exposure and 0.5 mSv/a via inhalation of radon. For future regulations in Germany, the approach is to restrict external exposure to 1 mSv/a and to restrict radon concentrations separately to a value of about 20 Bq/m³.

1. INTRODUCTION

Investigations of radiation exposure from radionuclides of natural origin in building materials have already been carried out in Germany [1–3]. However, since most of the measurements were made selectively (for instance, in cases of suspected enhanced radionuclide concentrations or NORM used as additives) and published in terms of pooled values with imprecise descriptions (such as ‘concrete’), they cannot be considered to be representative of the overall situation in Germany today. Moreover, the regulatory framework is currently being revised on the international scale. The revision of the European Council Directive 96/29/Euratom will include for the first time specific regulations on building material [4]. The European Economic Community 'Construction Products Directive' of 1989 [5], too, involves current efforts made towards unique measurements and evaluations of natural radioactivity in building materials in Europe. In either case, the recommendations contained in the European Commission (EC) publication 'Radiation Protection 112' (RP 112) [6] is the basis of the discussion. The main feature of this recommendation is to restrict external gamma exposure resulting from building materials to values below 1 mSv per year. This recommendation is being transferred into national legislation in Austria, Luxembourg, Poland and Finland, partly involving modifications concerning radon exposure. However, no such regulation exists in Germany up to now.

It is therefore important to have at hand up to date data on radionuclide contents and resulting radiation exposures, including information on the significance of the contribution of NORM used as additives to building materials. The information is needed to evaluate the consequences of the upcoming new regulations, taking into account the existing regulatory framework on NORM in Germany as well as recommendations on the control of exposure to radon in homes. Regulations on the use of NORM as an additive to building materials already exist in Germany. They are part of the German Radiation Protection Ordinance (RPO) of 2001 [7]. Concepts are needed for a common approach to the management of natural radioactivity in building materials including NORM on an international and national level. In Germany, care should be taken to ensure that the respective national regulations:

(a) Do not fall short of the protection level provided by the current NORM regulations that, in contrast to RP 112, explicitly consider radon exposure;
(b) Are consistent with the recommendation of the BfS, regarding radon in homes, of a target value of 100 Bq/m^3, which is consistent with a recent WHO recommendation [8];
(c) Do not impose undue restrictions on widely used building materials and the utilization of industrial residues.

This paper describes the current situation concerning the use of NORM as an additive to building materials and, in Section 2, the existing legislative basis. Section 3 presents the results of our newest investigations into radiation exposure from radionuclides of natural origin in building materials, including those originating from NORM additives. In Section 4, a possible regulatory approach is presented that is in line with the recommendations of RP 112 and the requirements listed above. Section 5 contains some conclusions.

2. NORM IN BUILDING MATERIALS IN GERMANY — REGULATORY REQUIREMENTS AND USE

The RPO [7] comprises a ‘positive list’ of materials underlying radiological surveillance (for example, waste rock, sludge, sands, slag and dusts of different origin) that are suited for building purposes and have been used for such purposes in the past. An example is so-called red mud, a residue from the production of aluminium oxide and used as an additive in the production of bricks in the past. Another example is residues such as sludge from the treatment of drinking water showing enhanced levels of ^{226}Ra and ^{228}Ra, although the latter are not currently regulated in the RPO. In the past, large volumes of slag from copper ore smelting have been used either directly as building material or as additives. In order to ensure that a dose level of 1 mSv per year is not exceeded with regard to the protection of members of the public, the RPO requires that the use of such residues in building materials be restricted in terms of the content and activity concentration of the building material. A general activity concentration surveillance limit of C = 1 Bq/g has been set, referring to the sum of the activity concentrations of those radionuclides of the ^{238}U and ^{232}Th decay series showing the maximum values within the respective series:

$$C_{U238_{\max}} + C_{Th232_{\max}} \leq C \tag{1}$$

In the case of more than 20% NORM residues being added to building materials for house construction, the surveillance limit C is lowered to 0.5 Bq/g. If the surveillance limits are exceeded it is also possible to show compliance with the dose criterion of 1 mSv per year by a dose assessment using realistic exposure scenarios and parameters. According to the RPO and the dose model underlying the above mentioned criteria, external exposure by gamma radiation as well as inhalation of radon and its progeny have to be taken into account.

The NORM regulations are based on information on the use of NORM that partly date back to the 1970s. Our study therefore included inquiries on the use of possibly relevant materials in Germany. Currently, the use of NORM as an additive to building materials is restricted to a few products. These are mainly certain kinds of concrete that make use of slags such as those from the steel industry (the activity concentration in blast furnace slag is 0.04–0.3 Bq/g ^{226}Ra and ^{232}Th and 0.2–1 Bq/g ^{40}K). In contrast to that, phosphogypsum is not produced in Germany any more. Ash from current coal combustion has been shown to be of minor relevance due to its low activity concentration. Red mud is not used in building materials any more. To a certain extent, sludge from waterworks is used in the production of bricks or concrete, although the content is usually in the range of only 1–2%. The activity concentrations range from 1 up to about 10 Bq/g.

NORM does not play a significant role in building materials today, although the extent to which this may be a consequence of the regulations of the RPO of 2001 is unclear. Nevertheless, the few materials to which NORM is added are precisely those used in large volumes in the construction industry. In the case of brick, the materials are known for their relatively high radionuclide content anyhow. The use of slag clearly increases the activity concentration in concrete. That is why our study was aimed at the establishment of an overview of the situation regarding natural radioactivity in building materials as a whole, including NORM.

3. THE MEASUREMENT PROGRAMME

The measurement programme was installed in collaboration with the German Building Material Association. This collaboration ensured the representativeness we strived for, a high degree of market coverage and also access to information we were interested in about the compositions and origins of the compounds of the 120 samples. Only samples of materials for indoor application were selected. Owing to the wide diversity of concrete compositions in use, with, for example, a possible slag sand content of the cement of up to 80% (CEM III/B), representativity for concrete products was not achievable.

3.1. Measurement methods

3.1.1. Radionuclide content

The activity concentrations of the samples were measured by gamma spectrometry with HPGe detectors and analysed according to a German standard measuring method for natural samples [9]. The samples were crushed to a grain size of about 3 mm, dried at 105°C for 24 h and enclosed in air-tight and especially radon-tight containers. To reach equilibrium between ^{226}Ra, ^{222}Rn and the measured short-lived progeny, these containers were stored for at least 21 d.

3.1.2. Radon

A standardized measuring method for radon exhalation rate is still lacking in Germany. Up to now, most of the measurements were made by electrostatic capture of the ionized alpha emitting progeny and subsequent time resolved alpha spectroscopy inside a half sphere located on top of the sample [10–12]. This closed chamber and accumulator method [13] is widely accepted by the industry concerned and by the scientific community. However, it should not be forgotten that the results depend heavily on the measuring conditions (for instance, the tightness of the half sphere on a porous material, like aerated concrete) and the interpretation (for instance, the influence of the inner voids and surfaces of a vertically cored or honeycomb brick).

To avoid these influences and to take into account the fact that the samples were delivered in different sizes and conditions, two additional measurement methods were developed. In the first method, a closed chamber with enclosed sample [13], the saturated radon concentration inside a sealed sample container was measured with calibrated scintillation cells to determinate the emanation rate of the crushed samples. The exhalation rate was subsequently calculated from the material properties with the boundary condition of a diffusion length greater than bulk material size, which is in good agreement with measurements of the diffusion coefficient of building materials [11].

The second method developed was an open chamber method. The sample container was well ventilated and the low steady-state radon concentration of the ventilated air was determined using the two filter method [14]. This method is based on the measurement of radon decay products built up in a cylindrical vessel. The measuring system used here is modified by a special aerosol dosing system allowing lower detection limits compared with conventional systems. Due to the optimized design of the sample and measuring container, as well as the aerosol generator, radon concentrations down to less than 1 Bq/m^3 could be measured with an acceptable degree of uncertainty.

A comparison of the results showed good agreement between the methods. Therefore, the results from both methods were used for the interpretation.

3.2. Results

The results of the activity concentration measurements are summarized in Table 1. Overall, the results are in fair agreement with previously published data. However, the radium concentrations of bricks from the eastern part of Germany were remarkably lower than expected from surveys that had been carried out in the late 1970s. This could have resulted from the reduction of the use of red mud

TABLE 1. MEASURED ACTIVITY CONCENTRATIONS IN BUILDING MATERIALS

	Number of samples	Activity concentration (Bq/g)								
		^{40}K			^{228}Th			^{226}Ra		
		Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Gypsum products	5	<0.020	0.120	0.051	0.0016	0.0058	0.0033	0.0038	0.013	0.0086
Lime sand bricks	3	0.035	0.180	0.115	0.0028	0.0072	0.0063	0.0041	0.010	0.008
Mineral wool	7	0.049	0.350	0.155	0.0047	0.064	0.022	0.016	0.080	0.035
Bricks	27	0.470	1.200	0.719	0.037	0.098	0.057	0.038	0.063	0.048
Cement and mortar	11	0.135	0.380	0.230	0.011	0.021	0.015	0.011	0.035	0.022
Tiles and ceramics	5	0.295	0.620	0.429	0.039	0.097	0.062	0.067	0.110	0.088
Ceramic raw materials	15	0.065	1.700	0.664	0.020	0.135	0.054	0.015	0.115	0.046
Aerated concrete	10	0.097	0.350	0.193	0.0048	0.019	0.012	0.008	0.026	0.018
Mortars	7	0.120	0.310	0.226	0.006	0.031	0.019	0.011	0.053	0.027
Plaster	19	0.012	0.220	0.075	0.0009	0.031	0.006	0.002	0.022	0.0077
Floor screed mortars	5	0.210	0.295	0.213	0.011	0.034	0.017	0.011	0.026	0.015
Lightweight concrete	7	0.710	0.850	0.845	0.028	0.083	0.049	0.027	0.098	0.062

Note: Mean values are arithmetic means.

as a colouring material and the closure of several minor pits that delivered clay with relatively high radionuclide concentrations.

Most radon exhalation rates are considerably less than 0.001 $Bq \cdot m^{-2} \cdot s^{-1}$, leading to estimated indoor radon concentrations of no more than 20 Bq/m^3 (see Table 2). The model room described in RP 112 was taken as a reference, with an air exchange rate of 0.5 h^{-1} and a diffusion coefficient of 1×10^{-7} m^2/s. Material-specific, realistic wall thicknesses and densities were used. The dose conversion coefficient used by UNSCEAR [15] was used here.

Table 3 gives a summary of the effective doses arising from external exposure to gamma radiation and internal exposure from the inhalation of radon. The contribution from external exposure was calculated according to the method described in RP 112, based on a standard room model. With regard to the effective dose caused by inhalation of radon and its short lived decay products, it should be noted that the use of the newly recommended dose conversion coefficient of the ICRP [16] leads to doses that are about 25% higher than those given in the third column of Table 3. The investigations showed that for some traditional building materials such as brick and light-weight concrete, the effective doses may be as high as 1 mSv/a via the external exposure pathway and 0.5 mSv/a via the inhalation of radon. These are moderate values. However, the use of different raw materials or materials with a higher NORM content could lead to considerably higher values.

4. A PROPOSAL FOR A FUTURE REGULATION IN GERMANY

The German NORM regulations and the recommendations of RP 112 reflect different regulatory approaches, in that:

TABLE 2. ESTIMATED CONTRIBUTIONS TO INDOOR RADON EXPOSURE FROM BUILDING MATERIALS

	Contribution to radon concentration (Bq/m^3)	Possible contribution to dose (mSv/a)
Gypsum, lime sand bricks, mortar, plaster, aerated concrete, tiles	0–4	≤0.1
Floor screed mortars	0–8	≤0.2
Bricks	0–12	≤0.3
Lightweight concrete, concrete, cement	0–18	≤0.4

TABLE 3. EFFECTIVE DOSES FROM EXPOSURE TO BUILDING MATERIALS

	Annual effective dose (mSv)	
	External exposure, gamma radiation	Internal exposure, inhalation of radon
Gypsum products	<0.3	<0.02–0.02
Lime sand bricks	<0.3	0.02–0.05
Mineral wool	<0.3	Not measured
Cement	<0.3	0.02–0.05
Tiles/ceramics	<0.3	0.002–0.005
Aerated concrete	<0.3	0.02–0.15
Mortars, plaster	<0.3	<0.02–0.02
Floor screed mortars	<0.3	<0.02–0.05
Bricks	0.3–0.9	0.02–0.30
Lightweight concrete	0.3–1.0	0.1–0.45
Concrete	0.3–1.0	0.1–0.5

(a) The 1 mSv per year criterion refers to the building material as a whole in RP 112 but to the contribution from NORM additives only in the German regulations;

(b) Radon is taken into account only implicitly in RP 112, whereas it is explicitly considered and dosimetrically evaluated in the German NORM regulations;

(c) ^{40}K is taken into account in RP 112, while the German NORM regulations refer only to the radionuclides in the uranium and thorium decay series.

It is obvious that these approaches are not completely compatible. A possible future abandonment of the current German NORM regulations of the RPO concerning building material in favour of a specific regulation based on the RP 112 approach may, at least theoretically, lead to a deterioration of protection against radon exposure. Therefore, it appears to be prudent to explicitly take radon exposure into account. If the effective dose is restricted to 1 mSv per year, the protection level will generally be at least as good as that provided by the current NORM regulations, which restrict only the contribution of NORM additives to 1 mSv per year. Together with the contribution from the original raw material, exposures of 2 mSv per year or more cannot be excluded currently.

Concerning radon exposure, there is another aspect that needs to be considered. The reason why no specific criterion on radon was established in RP 112 is the assumption that the design level of 200 Bq/m^3 of the EU recommendation on radon [17] for new buildings is unlikely to be exceeded if the requirements for external exposure are fulfilled. However, BfS is recommending a target value of 100 Bq/m^3, corresponding to the lower level of the 100–300 Bq/m^3 range recommended in the Radon Handbook recently published by the World Health Organization (WHO) [8]. Since the main cause of radon in homes is the underlying soil, the contribution from building materials should be small compared with the 100 Bq/m^3 target value recommended by the BfS. We consider a value of 20 Bq/m^3 as a suitable reference value for the contribution from building materials. Considering the results of our investigation, the likelihood of concentrations from the building material in excess of that value is low, at least if the contribution from NORM additives is controlled. However, it is possible that the contribution of building materials to the indoor radon concentration considerably exceeds 20 Bq/m^3 in the case of air exchange rates that are lower than the assumed value of 0.5 h^{-1}, for instance, as a result of measures to minimize energy consumption.

It is clear from our data as well as from those of a variety of previous investigations that the total effective dose from external exposure and radon inhalation may easily exceed 1 mSv per year for important bulk building materials such as concrete and brick, even without NORM additions. Since there are, on a large scale, no alternatives to these construction materials in Germany, the aim of radiation protection measures can only be to avoid extreme exposures and to restrict the addition of NORM on the basis of either an additional, independent radon criterion or a higher dose level. The latter option was chosen in Austria, where a dose level of 2.2 mSv per year was defined for the sum of external and radon exposure. We suggest that an independent radon criterion be introduced instead, for two reasons:

(i) A common dose criterion makes the dose contributions from radon and external gamma radiation interchangeable. This could lead to situations where the nationally defined dose criterion is fulfilled while the requirements according to RP 112 are not, with respective consequences on the export of such building materials.

(ii) The coefficient for the conversion of radon exposure into effective dose has changed several times in the past. The definition of a separate radon concentration level makes things independent of such developments.

Our recommendation for future radiological regulations on building material in Germany, therefore, is to apply both a dose criterion of 1 mSv per year according to RP 112 in order to restrict external exposure and, in parallel, a reference value of 20 Bq/m^3 for the indoor radon concentration to restrict exposure to radon from building materials.

5. CONCLUSIONS

NORM is currently not used to a large extent in building materials in Germany. Radiation exposures due to additions of NORM to building materials are confined to those giving doses of less than 1 mSv per year. The contributions of the traditional raw materials themselves are generally also lower than 1 mSv per year, as far as only external exposure to gamma radiation is concerned. This situation is to be regarded partly as a consequence of the new regulations on the use of NORM in the German Radiation Protection Ordinance and partly as a consequence of changing raw materials. Under these circumstances, future regulations on radioactivity in building materials are not likely to cause additional restrictions on the use of traditional building materials in Germany including NORM additives. If, according to the new WHO recommendations on indoor radon, a target value of 100 Bq/m^3 is envisaged, an additional limitation on the contribution of building materials to the indoor radon concentration appears to be reasonable. We consider a value of 20 Bq/m^3 as being adequate for Germany, applied independently of the restriction of external dose according to RP 112.

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RADIOACTIVITY IN PHOSPHOGYPSUM AND THE RECOVERY OF SODIC SOILS IN THE BRAZILIAN SEMI-ARID ENVIRONMENT

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Abstract

Soil salinity in the Brazilian semi-arid environment is a common problem caused by incorrect agricultural practices allied to local climatic and soil conditions. The use of phosphogypsum (PG) to recover these soils is still of some concern because this material contains radionuclides of natural origin. An experiment was conducted to study the use of PG to reduce the salinity of two major soils from the Brazilian semi-arid region. The radionuclide content of the PG samples was previously analysed with gamma spectrometry equipment. Three dosages of PG were mixed with topsoil samples in greenhouse conditions and, after a reaction period and controlled irrigation, the samples were submitted to single extractions based on a widely used sequential extraction method. Radium isotopes and ^{210}Pb in the extraction fractions were analysed by alpha and beta counting. Most of the Ra isotopes and ^{210}Pb were found to be bound to the residual fraction and, to a lesser extent, to the exchangeable fraction owing to the small levels of carbonates, organic matter and Fe and Mn oxides. The ^{226}Ra concentrations in the PG were lower than those set by the US Environmental Protection Agency for allowing the use of PG in agricultural soils. Application of the PG did not enhance the ^{226}Ra activity concentration in the soils analysed. However, after 30 days of addition, PG may contribute to the concentrations of ^{228}Ra and ^{210}Pb in the exchangeable fraction. The PG promoted a satisfactory reduction of electrical conductivity in the soils studied and indicates the possibility of reclamation of these soils.

1. INTRODUCTION

Soil salinization and sodification is a natural consequence of environmental factors such as deficient soil drainage and slope, depth and constitution, evaporation, and rainfall, mainly in arid and semi-arid areas. When combined with inadequate irrigation and other agricultural practices, this process is intensified. Soil salinization and sodification are common in semi-arid zones such as the north-east of Brazil, but is of concern worldwide because it represents the first step towards desertification, which has negative social, economic and environmental implications. One option for the reclamation of soils affected by the accumulation of sodium and salt is a chemical reclamation using natural gypsum and phosphogypsum (PG).

Natural gypsum is composed of gypsite, while PG is a by-product of phosphoric acid production. PG is generated in large amounts when phosphate rock is acidified with sulphuric acid to produce phosphoric acid [1]. PG is mainly composed of calcium sulphate, but also has trace elements and impurities such as fluoride, rare earth elements and radionuclides from the uranium and thorium decay series [2, 3], which vary according to the origin of the material and the production process [4]. Owing to the occurrence of these radionuclides, PG may be regarded as NORM. During PG production, radioactive equilibrium is disrupted and the uranium and thorium isotopes tend to migrate to the phosphoric acid, while radium and lead isotopes migrate to the PG [5–9].

In Brazil, as in the rest of the world, PG is stockpiled in areas next to phosphoric acid plants and is used for economic benefit. The material has been evaluated in several studies on the economic, agronomic and radiological issues [5, 12, 17–24].

Studies have been conducted on the PG radionuclide content and its use in agricultural soils [11–13], with the objective of reducing the aluminium concentration and increasing the infiltration rate [14, 15]. Other reports have focused on PG as a fertilizer to supply calcium and sulphur to crops and its application to agricultural soils affected by salinization [16] or as an alternative for the reclamation of saline and sodic soils to reduce the concentration of exchangeable sodium [25, 26]. The use of PG to reclaim soils affected by excess sodium also might minimize the problems created by stockpiling of this material. Owing to its radionuclide content, PG might promote an increase in the concentration of these radionuclides in agricultural soils and consequently increase their uptake by vegetables. Therefore, a knowledge of their behaviour is important for understanding their potential effects on the environment.

Radionuclides in soil occur with various forms of binding, depending on environmental factors such as the climate, the soil characteristics (including the pH and the composition of the mineral and organic fractions) and the physico-chemical form of the radionuclide. A determination of the types of binding provides useful information on the mobility and availability of radionuclides. However, soil is a complex matrix and the determination of the binding is difficult. One option is the application of extraction or fractionation methods allied with analytical procedures. The most common methods are single or sequential chemical selective extractions, developed with the purpose of identifying reversible and irreversible mechanisms in soils, the possibility of migration of the radionuclide within the soil and on the environment, and the possibility of vegetable uptake [28]. The results present evidence for possible geochemical associations of the radionuclide with the soil components, but the sequential extractions may present some difficulties such as the readsorption of the element onto solid particles before the separation of the aqueous and solid phases, non-selectivity of the extractors (the attack of another fraction that is not the target of the extractor) and incomplete dissolution of the target phases [29]. Single extractions are methods of fractionation using a unique extracting agent, but they also present problems such as the extraction of the soluble fraction by an aqueous reagent and the possibility of an extractor influencing the extraction of elements that are not of interest. For these reasons, single extraction procedures are not suitable for many types of study.

However, despite the decades over which the methods of single and sequential extraction have been developed, studied and applied, no method has yet been considered as universally accepted. Many methods have been tested with the focus on the extraction of heavy metals in soil, but the one described in Ref. [30] has been widely used and adapted for the extraction of radionuclides [28, 29, 31]. In this method, several reagents are applied sequentially (each extractor is chemically more active than the previous one) for the following five fractions: exchangeable (F1), bound to carbonates (F2), bound to Fe and Mn oxides (F3), bound to organic matter (F4) and residual (F5).

The objective of this work was to evaluate the behaviour of radionuclides of natural origin (^{210}Pb, ^{226}Ra and ^{228}Ra) in soils amended with PG, using a modified extraction procedure adapted from that described in Ref. [30]. The potential of PG to reduce exchangeable sodium concentrations and electric conductivity as indicators of soil reclamation was also evaluated.

2. MATERIAL AND METHODS

Samples of PG were collected from a stack situated at Imbituba, a city in the Brazilian State of Santa Catarina. This material was produced by a phosphoric acid plant that used mainly igneous phosphate rock, but also sedimentary phosphate rock from Brazil and other countries. The samples were homogenized, sieved in a 2 mm sieve and stored in polystyrene containers of 300 cm^3 capacity. After hermetic sealing, the samples were stored for 30 d to allow equilibrium to be reached. The samples were then analysed by gamma spectrometry. The detection system used in the PG radionuclide analysis (supplied by Ortec) comprised a high purity germanium (HPGe) detector for low energy and low background radiation, with an efficiency of 20% and 4096 channels. The detector was calibrated using a ^{60}Co source. The ^{226}Ra activity concentration was determined from the average activity concentration of its progeny ^{214}Pb (by measuring the intensities of the 295.21 and 351.93 keV energy emissions) and ^{214}Bi (using the 609.32 keV energy emmision). The ^{228}Ra was determined from the activity concentration of its progeny ^{228}Ac (using the 911.07 and 968.90 keV energy emissions). The counting time was set at 86 000 s. The Maestro program from Ortec was used for obtaining and recording the data. The spectra obtained were analysed using the computer program Aptec-NRC.

Two soils of wide occurrence in the Brazilian semi-arid region were selected for this study. The first was sampled in the municipality of Belém de São Francisco, in the Brazilian state of Pernambuco, and classified following EMBRAPA [32] and FAO [33] classification systems as Fluvisol (FL). The sampling point was located at 38°57'52,80” W and 08°41'35,54”S on the irrigated perimeter of Manga de Baixo. The second sample was classified as Luvisol (LV) and was located on a farm at 39°03'59,15” W and 08°41'08,39”S. FL is a soil that occurs in wetlands associated with recent sediments of fluvial origin and, when formed in semi-arid environments, often contains soluble salts or exchangeable sodium, which are responsible for the limitations on its use in agriculture. LV is a shallow soil, typically occurring in semi-arid environments, with a large difference in the sand–clay relationship between the A and B horizons. It usually exhibits a stony character on the surface and high levels of exchangeable calcium and magnesium, and is also subject to salinization. Both soils had been cultivated with onion and rice and then were abandoned owing to the salinity. Twenty-four samples of 12 kg (12 samples of each soil) were collected from the upper 20 cm layer and an experiment with these soil samples was conducted in a greenhouse. The soils were analysed for fertility and electrical conductivity as described in EMBRAPA [34]. The soils were also subjected to X ray fluorescence (XRF) and particle size analysis to obtain the chemical composition and the percentages of sand, silt and clay.

The soil amendment experiment consisted of evaluating three situations: (i) no PG applied, (ii) the required amount of PG applied (single dosage), and (iii) twice the required amount of PG applied (double dosage). The required amount, *RP*, is determined from the following equation [35]:

$$RP = \left(ESP_i - ESP_f\right) \times h \times CEC \times 86 \times D_s \tag{1}$$

where

RP	is the required amount of PG (kg/ha),
ESP_i	is the exchangeable sodium percentage found from chemical analysis of the soil (%),
ESP_f	is the final exchangeable sodium percentage in the soil, stipulated as 10% of ESP_i (%),
h	is the depth of application of PG (0.20 m),
CEC	is the cation exchange capacity ($cmol_c/dm^3$),
86	is the molecular weight of calcium sulphate (g/mol),
D_s	is the density of the soil (kg/dm^3).

The value of ESP_i was calculated from the following equation:

$$ESP_i = \frac{Na \times 100}{CEC} \tag{2}$$

The soil samples were packed in individual 12 L buckets and drained by means of a hole in the bottom into which a 1 L polyethylene bottle was engaged. Each soil sample represented an agricultural ‘plot’, which received the required amount of PG as calculated from Eq. (1) in the three dosages (zero, single and double). The single and double dosages were, respectively, 16 and 32 t of PG per hectare for Soil FL and 18 and 36 t of PG per hectare for Soil LV. The PG was mixed into the 20 cm layer of soil along with 1 L/d of irrigation water. After 30 d, samples of 300 g were collected from each bucket, homogenized, air dried, ground on a porcelain melting pot, sieved with a 200 µm sieve and then submitted to quartering. Subsequently, approximately 5 g were removed and subjected to sequential extraction according to the method described in Ref. [29], with the following five extraction fractions: exchangeable (F1), bound to carbonates (F2), bound to Fe and Mn oxides (F3), bound to organic matter (F4) and residual (F5). A total acid attack with HNO_3, HF and HCl was conducted on each sample in a Teflon beaker on a hotplate until complete digestion. These extracting steps were done as single extractions, not sequentially, to avoid interference with the extractions caused by the reagents and for other reasons as stated in Section 1. Another modification to the method was a change in reagent to sample ratio from 15:1 to 10:1.

The radium isotopes and ^{210}Pb on the extract solution of each fraction were co-precipitated as $Ba(Ra,Pb)SO_4$ by adding H_2SO_4 and $BaCl_2$ to 1 L samples. The mixed sulphates were washed and dissolved with nitrilotriacetic acid (NTA). Barium–radium sulphate was reprecipitated by adding acetic acid until the pH reached 4.5–5.0, whereas lead remained in the aqueous phase. The aqueous and solid phases were separated and the $Ba(Ra)SO_4$ was purified by dissolution in an ethylenedinitrilotetraacetic acid disodium salt dihydrate (EDTA) in solution at pH 10. Then the sulphate mixture was again precipitated by acetic acid (pH 4.5–5.0) and filtered. After one month of progeny ingrowth, ^{226}Ra was determined by gross alpha counting. Radium-228 was determined by beta counting, whereas the alpha particles of ^{226}Ra were stopped by a filter paper. Corrections were made if there was any contribution from the progeny of ^{226}Ra [36]. The radium isotope and ^{210}Pb counting was performed by alpha and beta counting using a Canberra Model S5XLB gas flow proportional detector. These analyses were performed by the Environmental Division laboratories of CRCN-NE/CNEN, Recife, which participate in inter-laboratory comparison exercises organized by IRD/CNEN, Rio de Janeiro.

3. RESULTS AND DISCUSSION

3.1. Soil characteristics

Soils show variations in physico-chemical properties, especially pH, electrical conductivity and exchangeable sodium percentage. These characteristics are used to classify the soil as saline, saline-sodic or sodic [37]. The results of the fertility and electrical conductivity (EC) analyses are given in Table 1. The EC, pH and ESP values indicate that both soils are saline-sodic soils. This situation promotes a reduction in available water due to the osmotic effect and a decrease in the infiltration rate. The phosphorus content in both is considered low by Brazilian standards and the pH values are not inadequate for the majority of crops cultivated in the region (rice and onion), with the soil classified as weakly acid. All the other exchangeable cations are at high values [38].

The results of the XRF analysis and particle size analysis (percentages of sand, silt and clay) conducted on samples of PG and soil are given in Table 2. The soils have a low content of Fe and Mn oxides and a high content of SiO_2 and Al_2O_3. Soil LV also exhibited a high level of SO_3 (50.98%) and CaO (40.478%). The PG results indicate that it has a high content of phosphate as P_2O_5 and also of oxides as Fe_2O_3, CaO, Al_2O_3, and SiO_2. The particle size analysis revealed that the soils have a high silt content and a low clay content, probably as a result of the presence of salt, which has the capability of dissipating clay and promoting a breakdown of soil structures.

TABLE 1. CHEMICAL AND PHYSICO-CHEMICAL CHARACTERISTICS OF THE SOILS USED IN THE EXPERIMENT

	Phosphorus content (mg/dm^3)	Electrical conductivity (mS/cm) (1:5)	pH H_20 (1:2.5)	Cation exchange capacity (cmol$_c$/dm^3)							Exchangeable sodium (%)
				Ca	Mg	Na	K	Al	H	CEC	
FL	27	15.87	5.20	10.45	3.95	7.00	0.24	nd[a]	3.38	25.0	28.00
LV	9	83.34	6.30	16.25	20.75	7.83	0.40	nd	0.24	37.8	20.71

[a] nd: not detectable.

TABLE 2. COMPOSITION OF PG AND SOILS USED IN THE EXPERIMENT

	Composition (%)[a]		
	PG	Soil FL	Soil LV
SiO_2	62.860	74.801	0.638
SO_3	0.365	1.063	50.983
Al_2O_3	20.570	10.952	nd[b]
P_2O_5	3.419	nd	nd
Cl	1.267	4.332	nd
Fe_2O_3	5.777	2.301	0.802
K_2O	2.595	1.864	1.864
MgO	1.520	1.296	1.296
CaO	2.460	1.144	40.478
Na_2O	1.215	1.141	1.141
Sand	9.5	32.1	35.3
Silt	89.2	59.7	53.6
Clay	1.3	8.2	11.1

[a] Some minor constituents are not included.
[b] nd: not detected.

3.2. Radionuclide concentrations in the PG samples

The activity concentrations of ^{226}Ra and ^{228}Ra in the PG samples were 0.06–0.13 and 0.06–0.24 Bq/g, respectively. These values are below the 0.37 Bq/g ^{226}Ra concentration considered as a limit by the US Environmental Protection Agency for the distribution and commercial use of PG for agricultural applications [10]. In general, the PG samples exhibited average activity concentrations slightly below those reported in the literature by other Brazilian investigators [5, 19]. The range of ^{210}Pb concentrations in the PG samples was 0.484–2.216 Bq/g, higher than values reported in other studies. A comparison of the activity concentration data with those reported in other investigations in Brazil and elsewhere is given in Table 3.

3.3. Results of fractionation of the soils

The radionuclide concentrations in each extraction fraction are given in Table 4. The values for zero PG dosage for the two soils can be considered as the concentrations in natural soil with no PG addition. For the soil samples amended with PG, most of the radionuclides are found in the residual fraction (F5) and, to a lesser extent, in the exchangeable fraction (F1), regardless of the dosage applied.

3.3.1. Lead-210 in soils amended with PG

In comparing the results in Table 4 for ^{210}Pb in the exchangeable fraction (F1) in soil LV with those for the total soil digestion (bulk), it can be concluded that the percentage of binding increases slightly with the dosage — the binding percentages were 7%, 16% and 15% for zero, single and double dosages, respectively. The same occurs in soil FL, with corresponding binding percentages of 9%, 6% and 11%, but the percentage of recuperation for single dosage was considerably smaller than that for zero dosage. This may indicate that, after the 30 d of PG addition, certain levels of ^{210}Pb may become available for vegetable

TABLE 3. ACTIVITY CONCENTRATIONS IN PG, BY COUNTRY

Origin of PG	Activity concentration (Bq/g)			Ref.
	^{210}Pb	^{226}Ra	^{228}Ra	
Florida, USA	0.435–1.369	0.433–1.110	nr[a]	[22]
Canada	0.890	nr	nr	[14]
Australia	0.451	nr	nr	[14]
Spain	nr	0.336–1.220	nr	[8]
Brazil	0.047–0.894	0.022–0.695	nr	[5]
Brazil	0.136–1.163	0.122–0.940	0.124–0.273	[19]
Brazil	0.484–2.216	0.060–0.130	0.060–0.240	This study

[a] nr: not reported.

uptake or for association with soil solution and/or minerals and organic fractions of the soil. However, the solubility and mobility of lead in soils are low [39].

The amount of lead bound to the carbonate fraction (F2) was expected to be low or not detected, as the soils have small values of carbonate minerals and the binding of ^{210}Pb with carbonates is extremely low [39]. It was therefore not surprising to find that the percentage of lead bound to the carbonate fraction (F2) of the soils could not be detected. The same situation was observed for ^{210}Pb in the organic matter fraction (F4) — the amount was not quantifiable, which could have been the outcome of the low content of organic matter in those soils due to the lack of vegetable activity caused by soil salinization.

For soil FL, the concentration of ^{210}Pb in the Fe and Mn oxide fraction (F3) is below the detection limit for single dosage but above the detection limit for zero and double dosages. Moreover, the concentration is higher for double dosage than for zero dosage. In soil LV, ^{210}Pb was detected in the F3 fraction only for zero dosage. It has been reported that the occurrence of lead bound with the F3 fraction can be observed more readily when pH values are low and in soils with a considerable Fe and Mn content [40]. Although this does not seem to be the case in the present study, taking into account the results of the XRF and particle size analyses in Table 2, it should be noted that, while the levels of Fe and Mn oxides in soil LV (2% and 1.2%, respectively) are lower than those in soil FL (6% and 1.5%, respectively), both soils have low levels of these oxides. This can explain the low concentrations of ^{210}Pb found in soil FL and values below the detection limit in soil LV.

The values of ^{210}Pb in the residual fraction (F5) were 50–78% of the total ^{210}Pb content for soil FL and 48–61% for soil LV. The two soils before the addition of PG had approximately the same ^{210}Pb content in the residual fraction (60–64%).

3.3.2. Radium isotopes in soils amended with PG

Radium is an alkaline earth element and has a relatively high mobility in the environment. Among natural radionuclides, it is the element that has the greatest potential for transfer between different environmental compartments [41]. According to Ref. [42], the mobility of radium, and consequently the vegetable uptake, is reduced in soils with high concentrations of calcium and magnesium. Generally, the concentrations of radium in soil are extremely low and the geochemistry is controlled by specific adsorption reactions (inner sphere complex) or non-specific adsorption reactions (outer sphere complex) with the minerals. The complexation with anionic species in the soil (such as OH, Cl, F, SO_4 and HCO_3) and organic matter (such as amino acids, fatty acids, phenols and hydrocarbons, urea, sugars, and humic and fulvic acids) also occurs. The radionuclides may also form colloids or be absorbed into their surface [43]. Some

TABLE 4. RADIONUCLIDE CONCENTRATIONS IN EXTRACTIONS FROM THE EXPERIMENTAL SOILS AMENDED WITH PG

	Soil extraction fraction[a]	Activity concentration (Bq/g)					
		Soil FL			Soil LV		
		Dosage = zero	Dosage = *RP*[b]	Dosage = 2 × *RP*	Dosage = zero	Dosage = *RP*	Dosage = 2 × *RP*
^{210}Pb	F1	0.0045 ± 0.0002	0.0037 ± 0.0002	0.0053 ± 0.0003	0.00248 ± 0.00018	0.00530 ± 0.00052	0.0052 ± 0.0003
	F2	—[c]	—	—	—	—	—
	F3	0.00022 ± 0.00001	—	0.00074 ± 0.00003	0.00031 ± 0.00006	—	—
	F4	—	—	—	—	—	—
	F5	0.03115 ± 0.00045	0.04563 ± 0.00358	0.02327 ± 0.00028	0.01967 ± 0.00024	0.01710 ± 0.00019	0.01607 ± 0.00019
	Bulk	0.0484 ± 0.00042	0.05903 ± 0.0041	0.0459 ± 0.0026	0.0327 ± 0.0022	0.03306 ± 0.0027	0.03327 ± 0.0013
^{226}Ra	F1	—	0.00085 ± 0.00007	—	—	—	0.00085 ± 0.00006
	F2	—	—	—	—	—	—
	F3	0.000930 ± 0.00002	—	0.000706 ± 0.000005	0.00608 ± 0.00043	0.000908 ± 0.000064	—
	F4	—	—	—	—	—	0.030 ± 0.002
	F5	0.01489 ± 0.00011	0.02108 ± 0.00018	0.01394 ± 0.00011	0.01296 ± 0.00011	0.02018 ± 0.00019	0.01449 ± 0.00098
	Bulk	0.01619 ± 0.00184	0.02194 ± 0.00198	0.01471 ± 0.00135	0.01904 ± 0.00133	0.02109 ± 0.00198	0.01550 ± 0.00105

TABLE 4. RADIONUCLIDE CONCENTRATIONS IN EXTRACTIONS FROM THE EXPERIMENTAL SOILS AMENDED WITH PG (cont.)

	Soil extraction fraction[a]	Activity concentration (Bq/g)					
		Soil FL			Soil LV		
		Dosage = zero	Dosage = *RP*[b]	Dosage = 2 × *RP*	Dosage = zero	Dosage = *RP*	Dosage = 2 × *RP*
^{228}Ra	F1	0.00272 ± 0.00019	0.00335 ± 0.00023	0.00288 ± 0.0002	0.002206 ± 0.00015	0.00477 ± 0.00033	0.00388 ± 0.00028
	F2	—	—	—	—	—	—
	F3	—	—	—	0.007. 6 ± 0.00053	0.00103 ± 0.00007	—
	F4	0.00037 ± 0.00003	—	—	—	—	—
	F5	0.0547 ± 0.004	0.05158 ± 0.00094	0.03828 ± 0.00065	0.04134 ± 0.00076	0.0571 ± 0.0039	0.02106 ± 0.00147
	Bulk	0.06423 ± 0.00002	0.057923 ± 0.000017	0.04500 ± 0.00016	0.05717 ± 0.00399	0.06848 ± 0.00476	0.0306 ± 0.0021

[a] F1: exchangeable; F2: carbonates; F3: Fe and Mn oxides; F4: organic matter; F5: residual.

[b] *RP*: The amount of PG required to reclaim the soil, as determined from Eq. (1).

[c] —: Below the lower limit of detection (5×10^{-7} Bq/g for Pb-210, 3.8×10^{-6} Bq/g for Ra-226 and 1.8×10^{-6} Bq/g for Ra-228).

studies show that radium can be adsorbed into soils and sediments rich in iron oxides and hydroxides, kaolinite, illite, vermiculite and montmorillonite [44, 45].

The pH is one of the factors that strongly influence the mobility of radium in soil — in acidic conditions, radium becomes more mobile. This effect can be found in soils with low pH due to the presence of organic acids [43]. As the soils used in the experiments have pH values closer to neutrality than to acidity, it is expected that the radium concentration will be higher on non-labile fractions.

Brazilian soils generally have higher levels of thorium than uranium [46]. Therefore, it is common for them to exhibit levels of ^{228}Ra higher than those of ^{226}Ra. In a study of natural radioactivity in another Brazilian state in the north-east region, Rio Grande do Norte, it was found that the average ^{226}Ra concentration was 0.029 Bq/g, little different from the value of 0.0259 Bq/g reported by UNSCEAR for areas considered as ‘normal’ [47].

The radium isotope data obtained from the extractions conducted in this study show variations in behaviour:

(a) In the F1 extraction, ^{226}Ra was detected only for a single PG dosage for soil FL (almost 4%) and only for a double dosage for soil LV (5%).

(b) Radium was detected in the F3 extraction only for zero dosage (almost 6%) and double dosage (5%), with the addition of PG having no apparent effect.

(c) For both soils, the ^{226}Ra content in the F2 and F4 fractions was below the lower limit of detection, as was the case for ^{210}Pb.

(d) In the F3 fraction for soil LV, zero and single PG dosages gave extraction percentages of 31% and 4%, respectively, while a double dosage gave no detectable concentration of ^{226}Ra, despite the small content of Fe and Mn oxides in this soil.

(e) The ^{226}Ra concentration in the residual fraction showed similar values for both soils. Soil LV exhibited ^{226}Ra values of 68–95% of the total ^{226}Ra content. For soil FL, the proportion of ^{226}Ra in the residual fraction was 92–96%. PG appears not to increase the activity of ^{226}Ra in soils.

(f) The ^{228}Ra content in the exchangeable fraction of soil LV increased from about 4% for zero PG dosage to 6% for single dosage and to 12% for double dosage. The concentration of ^{228}Ra in this soil seems to increase in proportion to the dosage. The same did not occur with soil FL, for which the values increased only slightly with dosage (4%, 5% and 6%, respectively).

(g) In general, the addition of PG seems to increase the labile fraction, at least that of ^{228}Ra. As a result, the soil to plant transfer factor (TF) for ^{228}Ra should be higher in PG amended soils. Differences between the TFs for ^{226}Ra and ^{228}Ra have been observed previously [48], with the higher TF value for ^{228}Ra being ascribed to the association of each isotope with its respective parent radionuclide (^{238}U and ^{232}Th). Therefore the physical and chemical differences between minerals might result in differences in radium isotope availability for plants [39].

(h) The Fe and Mn oxide fraction seems to be more important for ^{226}Ra than for ^{228}Ra. The fraction of ^{226}Ra in the residual fraction (around 95%) is generally more elevated than that of ^{228}Ra (around 80%).

(i) While the F2 extractions did not show detectable concentrations of ^{228}Ra in either of the two soils, this radionuclide was detected in the F4 extraction of soil FL, but at a very low level of extraction (0.4%).

(j) The F3 extraction percentages for ^{228}Ra for soil LV were 13% for zero PG dosage, 1% for single dosage and below the lower limit of detection for double dosage, but this soil has very low levels of Fe and Mn oxides. This may suggest that PG causes a shift of ^{228}Ra from the F3 to other phases such as the residual and the exchangeable phases, or even to F4 on soils with a higher content of organic matter.

(k) For all the PG dosages, the concentration of ^{228}Ra found in soils was higher than the concentration of ^{226}Ra, as would be expected for Brazilian soils.

Although this study did not perform extractions from PG samples, studies have been conducted in Brazil of sequential extraction protocols applied to PG samples from other origins. These studies showed that the majority of the radium isotopes and ^{210}Pb are bound to the Fe and Mn oxide fractions (F3), but a portion of these elements are also found in the labile fraction (F1) [19, 23, 25]. These results are expected to be valid for the PG assessed in this study. It has also been concluded that, although these elements are found in PG, they are not associated with the calcium sulphate and therefore do not represent a threat to the environment [23].

3.4. Reclamation of saline-sodic soils

The values of EC in the experimental soils are shown on Table 5. Comparing with the values of the original soils (zero PG dosage), it can be seen that the EC after 30 d of PG addition decreased considerably. For both soils, single dosage gives the best value, indicating that a overdosage of PG may elevate the EC in those soils. The EC values for Soil LV after addition of PG indicate that it remains classified as a saline soil.

4. CONCLUSIONS

The ^{226}Ra and ^{228}Ra activity concentrations in the PG selected for this study are similar to those reported previously for other PG samples of Brazilian origin,

TABLE 5. ELECTRICAL CONDUCTIVITY OF THE EXPERIMENTAL SOILS

	PG dosage	Electrical conductivity (mS/cm) (1:5)
Soil FL	Zero	15.87
	Single	2.74
	Double	3.74
Soil LV	Zero	83.34
	Single	6.81
	Double	11.77

and are lower than those reported for PG originating from other countries. However, the ^{210}Pb concentrations are higher than values reported previously.

The addition of PG to two experimental saline-sodic soils indicates that, after 30 d, increases in the activity concentrations of ^{210}Pb and ^{228}Ra in the exchangeable fraction of the soil may occur, resulting in an increase in the transfer of these radionuclides to plants. Nevertheless this trend should be verified and quantified by soil to plant transfer factor experiments. The addition of PG appears not to increase the activity concentration of ^{226}Ra in the soil.

The activity concentrations of radium isotopes and ^{210}Pb were below the lower level of detection in the F2 (carbonates) and F4 (organic matter) extractions of the soils, owing to the low content of carbonates and organic matter in the soils of the experiment. These radionuclides are bound in the F5 (residual) and F1 (exchangeable) fractions, with only moderate concentrations in the F3 (Fe and Mn oxides) fraction, again as a result of the small amount of this mineral phase in the soils under investigation. Other soils with different compositions could exhibit different radionuclide behaviour.

Compared with the values measured in the original soils, it was found that the EC had decreased considerably 30 days after the addition of PG. The best EC values for both soils were obtained from a single dosage of PG (the calculated ‘required amount’), indicating that an overdosage of PG may elevate the EC in these soils if it contains significant concentrations of Ca and Mg.

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RAPPORTEUR SUMMARY OF TOPICAL SESSIONS 4 AND 5

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1. INTRODUCTION

The papers presented in Topical Session 4 can be broadly assigned as relating to the energy sector and the environment. The papers presented in Topical Session 5 were generally related to agriculture and construction uses of NORM containing products and residues.

2. ENERGY

As is the case in any industry utilizing NORM or impacted by incidental NORM, characterization of NORM in terms of source materials, distribution of radionuclides during the process, and their representation in products and residuals is critical to evaluate the need for regulation to protect workers and the public. Several papers addressed the NORM characterization issue and its implications.

In the Norwegian petroleum industry, NORM was characterized and low doses were estimated for both offshore workers (<0.5 mSv/a) and onshore workers (<0.05 mSv/a). Waste streams were also categorized and disposal solutions were presented, but with significant gaps that will require further research and development.

A particularly interesting mining site in the Democratic Republic of Congo produces both copper and cobalt, but also includes a history of mining and processing of uranium ore. Although the present activities are related to metal production, the energy related history of the site presents NORM challenges that range from management and engineering to public notification and education.

Another paper discussed evaluation methods for NORM containing adsorbents in a Japanese natural gas industry. The authors developed and compared methods to predict NORM (^{210}Pb) loading in adsorbents. The goal is to change the adsorbent before concentrations exceed the exemption level, thus avoiding a costly disposal situation. It is intriguing that the exemption level, not the adsorption capacity, is driving the frequency with which the adsorbents are being exchanged.

3. ENVIRONMENT

A unique paper about the characterization and management of the environmental NORM burden provided insight into a frequently overlooked area of industrial impact. The distribution, interaction, mobility and bioavailability of NORM in ecosystems contribute to the complexity of evaluating the impact on non-human receptors. In addition, the long half-lives of many NORM radionuclides, the preponderance of alpha emissions, and direct contact of NORM with biota create enduring and proximate external and internal exposure conditions.

In such conditions, and considering non-human dose receptors, traditional anthropocentric risk models are not appropriate and standard dose assessment techniques are untenable. The first indication of radiation impact is at the cellular level where cytogenetic evaluation methods are more appropriate. Environmental detriment can be determined using a multi-stage evaluation process, but there are significant knowledge gaps that must be filled to implement it.

Environmental modelling was used in a study of the dispersion of NORM (^{226}Ra) in a marine system, specifically releases to the Mediterranean Sea from discharge points in Morocco and Spain. The model can be used in planning future NORM releases (which involves legal issues that were not addressed) or in emergencies as a rapid response model.

4. AGRICULTURE

All the papers dealing with agriculture investigated ingestion pathway doses from the use of phosphate industry products and co-products.

One product of the phosphate industry is defluorinated dicalcium phosphate, which is used as an animal feed supplement. The paper reported results of the NORM fluxes during processing as well as NORM uptake (^{210}Pb and ^{210}Po) and distribution in the organs and tissues of poultry with subsequent ingestion dose to humans. This pathway yields doses of <20 µSv/a, which is well below current public health guidance levels.

Two other papers described the application of phosphogypsum to soil in Brazil. Phosphogypsum has been known to be beneficial in the remediation of sodic soils for many years, but regional solutions require investigation of indigenous soil types and phosphogypsum sources. Locally produced phosphogypsum was applied for sodic soil treatment and resulted in no enhancement of ^{226}Ra in soils, but ^{228}Ra and ^{210}Pb require further research. In related research, soil to plant transfer factors were low in clayey and sandy soils for the six NORM species investigated.

5. CONSTRUCTION

Phosphogypsum was used in the fabrication of wall panels and evaluated for external and inhalation dose pathways in a test house in which the panels were used on all exposed surfaces. Both the external dose and radon concentration as projected from measured data were within current limits and guidelines. Specifically, the estimated dose from external exposure was <0.3 mSv/a and the radon concentration was <200 Bq/m^3 in the closed structure.

In the titanium dioxide industry, co-products were characterized for their content of radionuclides of natural origin. Red gypsum was determined to be safe for use in cement production and enhanced the physical characteristics for use of cement in construction.

NORM residues were characterized for a wide variety of building materials commonly used in Germany. Construction materials that incorporate NORM residues were compared with traditional building materials in terms of contents and dose implications. NORM industry residues overlap the range of natural background NORM content in traditional construction materials. In this study, it was revealed that doses received via external gamma exposure and radon exposure from the use of building materials incorporating industrial NORM residues are not likely to exceed those received from the use of traditional materials, possibly because regulations to control radioactivity in building materials were already in place.

6. SUMMARY LESSONS

The IAEA provides international guidance on the appropriate levels of regulation (or non-regulation) of NORM industries and their residues. Non-regulated NORM is simply ‘material’ and there is therefore no need for the separate term ‘TENORM’. All NORM residues should be managed using ‘good practice’. However, it must be realized that good practice is situation specific and cannot be generically applied across industries. This concept is applied in the International Association of Oil and Gas Producers NORM Management Guideline presented during the day.

Industry needs to provide input, as it does in the IAEA Safety Reports, and it needs simple ‘how to’ guidance. This guidance can be in the form of good practice, but there is a hesitance to use the term ‘best practice’ as it may be site specific.

The scale of NORM problems can vary significantly between industries but also within an industry as evidenced by the various characterization studies presented in the two sessions. Activity concentration and volume determine the optimum disposal strategy for NORM residues consisting of characterized individual radionuclides with known emissions and half-lives.

Residues of large volume and low activity concentration may be considered for use as co-products rather than disposal, as evidenced by the safe use of phosphogypsum as an agricultural amendment, as a treatment for sodic soils and as a construction material. In the Netherlands, even some material that must be stored will be used after allowing sufficient time for decay. In contrast, small volumes of material with a high activity concentration are disposed of in long term containment as demonstrated again in the Netherlands (isolation, control and monitoring) and in the Norwegian petroleum industry.

Other non-radiological properties may limit the options for use or disposal. Also, attention was drawn to measurement problems, the importance of competent radiation protection personnel on NORM sites, and how consideration of volume and activity concentration was used to exclude the household use of NORM from legislative control.

Good practices are dynamic as industries develop and new technologies modify NORM distribution in processes, products and residues. Consequently, periodic review and revision of good practices are necessary.

Competency and community awareness are keys to sustainability of any industry and in maintaining safe operating conditions. The characterization of the copper/cobalt mining site in the Congo wisely pointed out the critical importance of worker training programmes (capacity building) and increasing public awareness of radiological hazards (stakeholder engagement).

In conclusion, a pragmatic, risk-based approach to safety should be adopted when examining disposal options for NORM industries.

CONCLUDING SESSION

Chairpersons

A. POFFIJN
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Focus On Uranium: Invited Paper

URANIUM MILL TAILINGS REMEDIATION IN CENTRAL ASIA

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Abstract

Uranium ore is a naturally occurring radioactive material which is often regarded as something separate to NORM due to its place at the front end of the nuclear fuel cycle. Uranium mining and processing was a significant industry in the Central Asian countries of the former Soviet Union. When the Soviet Union broke up in 1989 these countries gained their independence but the uranium mining industry now had to try and survive in a new economic environment. In Tajikistan and Kyrgyzstan this proved too great a challenge. Production stopped and sites were simply abandoned with little or no attention paid to remediation. Skilled personnel departed and both physical and regulatory infrastructure decayed. Consequently, the legacies of the former times remained throughout Central Asia to become an issue of considerable concern to many. The sites were generally uncontrolled and the NORM residues from the mining and processing were a source of environmental contamination which also threatened public health in a number of ways. In recent years there has been considerable activity by a number of international agencies and Governments working towards solutions for these issues. Much of the effort has been undertaken by the IAEA and this paper describes the original situation, the development of remediation strategies and the various remediation related projects, their outcomes to date, and plans for the future in both the political and scientific arenas.

1. INTRODUCTION

One of the most commonly occurring forms of NORM is uranium ore and yet for many years it has been regarded as something exceptional since it is the source material for nuclear fuel and thus right at the front end of the nuclear fuel cycle. In recent times there has been a move to diminish the distinction between uranium ore and other ‘more conventional’ types of NORM; in part, this has been to avoid unnecessary duplication of rules and regulations — this duplication seems to be less than optimal when it is realized that the safety standards to be applied to the production, management, use and residue disposal of NORM are the same regardless of whether the material is from a uranium mine or any other NORM source. Also this effort has been applied to try and make the public awareness of the real dangers of uranium mining more in line with the real risks, especially when compared to other ‘more acceptable’ NORM-based industries.

The uranium mining industry has had a chequered past with several of ‘boom and bust’ cycles already in its relatively short modern life. Modern uranium mining only began in earnest after 1945 when the major world powers were keen to obtain supplies of the seemingly rare mineral for use in fabricating nuclear weapons. Within a few years, as the requirements of the weapons industries were met, the demand for uranium diminished; only to be reinvigorated by the push to develop nuclear power which in turn required uranium for fuel. Then that demand slowed and then the ‘cold war’ came to an end — a double blow to the uranium mining industry which by the early 1990s had entered a severe decline in activity. Nowhere was this more clearly evident than in several of the Central Asian republics that had been major uranium suppliers to the former Soviet Union. With the change to independent governments and free market economies the older uranium mines were no longer economic and so were abandoned. These legacy sites are scattered throughout the region and present an ongoing source of detriment to the environment and population in the areas where they are located.

As the Governments in the newly established Central Asian republics became more established they realized that these legacy sites were an issue that could not be ignored. Equally they assessed the situation that they no longer had the resources to deal with the proper remediation of such sites; neither in financial terms nor in adequate numbers of suitably skilled and experienced people to manage such works. Also in most cases the regulatory and institutional infrastructure to supervise such activity had been dismantled with the collapse of the Soviet Union and no suitable replacement was available.

It was at this time that the IAEA became involved — the various Central Asian republics became Member States of the IAEA and were thus able to access its services and facilities. A series of preparatory missions was undertaken by IAEA staff and consultants throughout the region to collect basic data and make an initial appraisal of the situation. The culmination of all these activities came in 2003 when the Member States of Kazakhstan, Kyrgyzstan, Tajikistan and Uzbekistan all applied to for assistance under the IAEA’s technical cooperation (TC) programme. Among the wide range of topics under consideration for TC assistance, it was noted that each of these Member States had a request for a project relating to uranium mining and ore processing legacy sites.

Given this common situation within the four countries the decision was taken by the IAEA to design and implement a regional project within the

2005–2007 TC cycle for the four countries, entitled Safe Management of Residues from former Uranium Mining and Milling Activities in Central Asia. The rationale for establishing the regional project was that all four Member States had similar issues relating to abandoned uranium mining and processing facilities across a region with similar climatic conditions and covering the same geographic area. In addition, it was hoped that having such a project would encourage interaction and exchange of knowledge and experience between the four Member States; it was also hoped that working together would also strengthen the local institutions and improve resources. Finally, a regional project in this style would allow the IAEA to offer a more efficient and improved service to the region. The four participating Member States agreed and the project was begun in January 2005.

2. THE LEGACY SITUATION IN CENTRAL ASIA

The uranium mining activities of the former Soviet Union were widespread throughout the Central Asian Republics of Kazakhstan, Kyrgyzstan, Tajikistan and Uzbekistan. The majority of these operations began in the mid to late 1940s and were operated by State run companies with little or no consideration of the need to be profitable in a conventional economic way. However, when the break-up of the former Soviet Union came in 1991, only some of the sites were still in operation and many of these mining and processing sites ceased activity at that time. All too often there was little or no remediation of the site or the associated waste disposal facilities such as tailings dams and waste rock piles; this was a consequence of the downturn in economic activity, which meant many sites were simply abandoned. These legacy sites potentially contain a wide range of hazards to the environment and the population in the areas surrounding them. These may be physical (such as old buildings, open mine workings, pits and tunnels, and derelict buildings and machines); chemical (acid drainage from reactive waste, and old processing chemicals and residues); and/or radiological (such as tailings, unprocessed uranium-bearing ore, scale and sludge in old plants, and contaminated scrap metal).

The extent of the legacy issue has been estimated in a number of places but the most authoritative account is thought to be that in the end of project report for the original IAEA project. A summary of the data is given in Table 1.

TABLE 1. URANIUM MINING RESIDUES IN CENTRAL ASIA AND MONGOLIA

	Tailings sites		Mass of rock and low grade ore in waste dumps (million t)	Total in the country (million t)	Area of affected land (km^2)
	Number	RW mass (million t)			
Kazakhstan	3	246	37	283	52[a]
Kyrgyzstan	34	77	209	286	6.5
Tajikistan	10	55	115	170	3
Uzbekistan	1	60	13	73	3
Total, Central Asia[b]	48	438.5	374.1	812.6	64
Mongolia	2	0	6	6	1.5

[a] Excluding Semi Palatinsk.
[b] No data available for Turkmenistan.

3. THE PROJECT PLAN

The objectives of the regional project were (within each participating Member State):

(a) To develop a regulatory framework and decision making process to assess the radiological impact of radioactive residues at former uranium mining and processing sites;
(b) To evaluate the remediation works underway;
(c) To ensure international safety standards were being met;
(d) To develop a plan of action to minimize the impact of radioactive residues on the population and assist sustainable development.

These objectives also had to take into account the fact that two of the Member States (Kyrgyzstan and Tajikistan) also had national projects under the overall TC programme in the same time cycle that were related to issues of uranium mining residue remediation and safety. Obviously the work programmes of the various projects had to be checked and monitored to ensure that they remained integrated and complementary rather than overlapping and duplicating.

Five main types of activities were employed to achieve the objectives of the regional project. These were workshops, training activities, equipment supply, expert missions and scientific visits.

The largest component was a series of four workshops which were spaced more or less evenly throughout the project's life. In essence, each Member State was expected to host one workshop over the two year period. The workshops were structured so that over the project's life span they introduced the concepts and principles required to start planning the improvement and remediation of the existing situation. These topics also included planning activities, methods of data collection, how to design and implement monitoring and surveillance systems, how to justify and evaluate monitoring and surveillance programmes, and finally the importance of appropriate data assessment and reporting. In all the workshops it was emphasized that there is a need to employ suitable international standards in such work and much use was made of the relevant IAEA documentation. In particular, an IAEA Safety Guide on the management of residues from mining and processing of radioactive ores [1] and an IAEA Safety Report on the monitoring and surveillance of uranium mining and milling waste [2] were both used as standard texts.

The workshops were held in Dushanbe and Almaty (2005) and Tashkent and Bishkek (2006) at approximately 5–6 month intervals. During the workshops the participants were given opportunities to practice what had been presented in the workshops with both practical exercises and assignments being set by the organizers. Also excursions to legacy uranium mine waste sites in Tajikistan and Kyrgyzstan during two of the workshops (Tashkent and Bishkek) provided the opportunity for participants to hold discussions 'on the ground'. Everybody observing the same site at the same time provided the opportunity to discuss issues with a greatly reduced risk of any misunderstanding arising from an interpretation of the site description as provided by others, in a presentation for example.

The second project activity was the provision of on-site training for supervisory and technical staff involved in surveillance and monitoring tasks. These events provided an opportunity for participants to become familiar with the various items of equipment provided in the procurement part of the project, as well as reinforcing existing skills. It also enabled experts to discuss with participants the use and maintenance of their existing equipment. The prime objective was to upgrade the participants' ability to establish and maintain surveillance networks, for example monitoring ground and surface water around the legacy sites. The main items addressed were: the selection of sampling locations and appropriate sampling methodology; the selection of analytical methods; data recording and analysis; the reporting and archiving of data; and the use of data in preparation of remediation plans and for provision of assurance to the public. Throughout all these activities, the need for quality assurance and control was emphasized.

The third component was the procurement of some basic items of equipment to enable the upgrading of performance in surveillance and monitoring services in their respective countries. Equipment was supplied for both laboratory and field activities with a number of items supplied being deliberately common to all groups to facilitate the comparison of data — this was especially important in cross border monitoring discussions. There were also opportunities to provide items to address site specific needs in some instances.

The fourth component was the organization of scientific visits (SV) for some of the participants to observe what had been achieved elsewhere in the remediation of former uranium mining and processing facilities. The project established a good working relationship with the Wismut company in Germany. This organization is responsible for the remediation of all the former uranium mining and processing facilities in the States of Thuringia and Saxony in eastern Germany. The Wismut company inherited a mining legacy which included several open pits, about 1400 km of underground mine workings, 311 million m^3 of waste rock and 160 million m^3 of radioactive tailings. All of these residues were located in or adjacent to populated areas, thus the challenges faced by Wismut are comparable to those facing the participating Member States. The SV programme was organized so that two groups of personnel from regulatory organizations and two groups representing mine operators from each country spent about one week at the Wismut operation on each visit. The programmes included visits to both ongoing and completed sites to see examples of remediation activity; also briefing meetings with local authorities and the regulatory body; and opportunities to observe monitoring and surveillance activities actually being undertaken by Wismut staff.

The final component was to undertake a series of expert missions at intervals throughout the programme to provide ongoing support, encouragement and assistance to the participants. These missions also helped to facilitate liaison between the various groups involved in the countries and some cross border liaison as well. This last activity was very important as there were a number of other activities taking place in the region related to the remediation of uranium mining legacies being organized by a number of different agencies and organizations. With all the assistance agencies needing to optimize their allocation of resources, this liaison process was a very significant activity.

4. LIAISON ACTIVITY

The IAEA is not the only organization working on issues of remediation for legacy uranium mining activities in the central Asian region. A number of other organizations have also been undertaking assistance activities involving these legacy sites, and in some cases they had begun earlier than the IAEA. Perhaps the largest programme already running in 2005 was the Environment and Security Initiative (ENVSEC) being implemented in the Ferghana valley by a group of agencies [3]. The members of the group were the Organisation for Security and Cooperation in Europe (OSCE), the United Nations Development Programme (UNDP), the United Nations Environment Programme (UNEP) and the Science for Peace programme of the North Atlantic Treaty Organization (NATO). ENVSEC had several components, but one specifically dealt with matters of uranium mining waste safety and disposal. This programme contained elements of training and equipment supply which obviously were very similar to the activities within the IAEA project. In order to avoid any duplication of effort which would result in a sub-optimal application of combined resources to help with the resolution of these issues in the region, an informal system of liaison was implemented.

Another agency working in the region was the World Bank, which was implementing the Kyrgyz Republic Disaster Hazard Mitigation Project [4]. A significant part of this project involved the improved management and long term stabilization of uranium mill tailings in the area of Mailuu-Suu, western Kyrgyzstan. Again, the overall project plan contained components relating to technical training and the provision of surveillance and monitoring equipment which could have led to duplication of effort or procurement and thus less than optimal use of resources.

Another agency working on related issues was the Lawrence Livermore National Laboratory in the United States of America, which has been providing funds and coordinating resources for low cost, smaller scale remediation of uranium mill tailings sites at Kadji Say, Kyrgyzstan.

It was important to demonstrate to the Member States involved that information exchange and coordination of these various efforts were being put in place. Accordingly the IAEA incorporated liaison activities from the beginning of the project. For each workshop the IAEA extended an invitation to each of the other agencies to attend and make a presentation to update all parties on the progress of their respective programmes. Each agency was involved in at least one of the meetings. This has ensured that the participants could better understand how the various activities fitted together and showed the value of communication and liaison. Between workshops the various organizations exchanged information on equipment supply and training activities to ensure that the various elements of the overall activity stayed coordinated. This prevented unnecessary duplication of equipment supply and so enabled the range of items provided to be better coordinated. Details of the various training courses were exchanged, again to ensure that activities are complementary to the greatest extent practicable.

Throughout the initial project period and the subsequent extension into 2007–2008, these liaison activities have remained active. So much so that when the project was revised and recommended for implementation in a new format for 2009–2011 the existing liaison network proved extremely useful in facilitating organizations of various meetings relating to possible regional strategy formulation and preparation of conferences for potential donors.

5. PHASE 2 —RER 3010

As a follow-up to Phase 1, a new TC project RER 3010 entitled Supporting Preparation for Remediation of Uranium Production Legacy Sites has been implemented for the 2009–2011 programme cycle. The concept was to build on the knowledge and progress made under the previous project RER 9086 and to expand the activities to reflect a progression from site characterization through to remediation planning and implementation. Project RER 3010 comprises seven thematic work packages (WPs) designated WP1 to WP7. Each participating Member State has identified a ‘pilot site’ which will serve as a model for the appropriate work package activity. For example, Tajikistan has identified Degmay as their pilot site; it is planned that each of the following WP activities will be applied in the context of this site. The idea is that at the end of the project each Member State will have a template which can be used to apply to other uranium legacy sites.

In addition to the original four Member States (Tajikistan, Uzbekistan, Kazakhstan, and Kyrgyzstan), representatives of Romania, the Russian Federation and the Ukraine have joined the new project. The new project has been implemented to provide technical assistance to the countries in preparation of a national strategy of remediation of the so-called ‘uranium production legacy sites’.

The following is a brief description of each work package.

Work Package 1 (WP-1): Safety Assessment Methodology

The purpose of this activity is to acquaint participants in the project with modern safety assessment methodology, including the evaluation of the current situation (environmental contamination and human exposure) and also for long term impacts utilizing modern tools such as risk assessment models. A requirement for this WP is that for each of the chosen former uranium production pilot sites, all necessary input data for assessment will be collected by the national experts. The participants of the working group from the Member States have received training in methods for assessing the transport of radioactive substances in the environment, dose calculations and ecological risks estimation as a first activity under this WP. This was accomplished utilizing ECOLEGO software at the Facilia training facility in Sweden in January of 2010. Two tiers of training were provided, introductory and advanced. Each participating Member State was provided with a licensed copy of the software for its future use. The idea is to utilize this tool for safety assessment characterization of the hazards for each ‘pilot legacy site’ by the national experts under the methodical supervision of international skilled experts. As a result of the project implementation, each of the chosen pilot sites will have gone through a basic assessment, including analyses of the current situation and also of possible scenarios related to long term potential hazards. The assessment scenarios will include consideration of possible mitigation actions and remediation strategies for dose and risk reduction. The optimization of possible remedial actions will be included as an exercise as part of this WP for future remediation planning. A second meeting is planned in November 2010. In the interim, the participants are utilizing the pilot site data with the tools for presentation and discussion and further refinement in November.

Work Package 2 (WP-2): Enhancing the Regulatory Framework

The task for this work package is to review the current status of regulatory provisions relevant to remediation planning and implementation as it relates to compliance with international standards and good practice. Special attention will be given to the licensing of remediation activities. This will be accomplished through the provision of international experts who will consult the regulatory bodies on the application of new approaches proposed in ICRP and IAEA documents for establishing dose constraints, criteria for release of the sites from the regulatory control, criteria to evaluate the effectiveness of the remediation, requirements for long term monitoring, and surveillance and maintenance at the legacy sites.

With the help of international experts, the goal is for important regulatory documents, as well as some site specific recommendations on regulatory provisions, to be improved or developed. This will be accomplished by evaluating the pilot site to see if certain requirements exist, such as: is there a requirement for site monitoring and reporting to the regulator? It is a bottom-up approach which will be done by the regulator with assistance from international experts. This is the approach which was successfully used by the Norwegian Radiation Protection Authority (NRPA) in the Russian Federation. In fact, NRPA has entered into a series of bilateral agreements with three of the Member States in Central Asia to implement this same type of activity. IAEA and NRPA will work closely together in this area to maximize resources and impact. A bilateral meeting was held in Drammen, Norway in December 2009 in which the IAEA participated for the purpose of coordinating efforts. A workshop for WP-2 is planned in Kiev in September 2010.

In addition, an Internal Forum for Regulatory Supervision of Legacy Sites (RSLS) has been established by the IAEA. The first technical meeting will be held in Vienna in October 2010 and will focus on Central Asia uranium legacy sites. While not specific to Project RER 3010, it is certainly relevant and project participants will be involved.

Work Package 3 (WP-3): Establishing Site specific Monitoring Programmes

Based upon results obtained from the previous project (RER 9086) and discussions with experts from Member States, there is a recognized need to further develop or establish site specific monitoring programmes associated with the uranium production legacy sites. Data gathered from such programmes would provide additional input to a safety assessment and would provide a justification of the remediation strategies. Currently, as has been shown by national presentations and also from the experience of the previous projects in the region, the environmental monitoring programme designs at many of the legacy sites are not optimal. The goal of WP-3 is to review the existing monitoring plans and activities for the pilot site to evaluate and enhance them, or establish them where they do not exist. The main purpose of the activities planned in WP-3 is to assist Member States in optimizing methods of data collection, to refine and develop sampling plans and to improve data management and interpretation. Once the existing plans are reviewed, international experts will work with regulatory experts to help develop guides for environmental monitoring including air, soil and water. This WP will include an activity to evaluate indoor and outdoor radon concentration at the former industrial sites and surrounding inhabited areas of the pilot site where appropriate. This activity is a crucial part of the decision making process in remediation planning. Participants in this WP will have an opportunity to be acquainted with experience and progress achieved in establishing monitoring in other countries involved in the project. Examples from countries from outside the region will be presented as well. A workshop in Pecs, Hungary is planned for August 2010 at a former uranium mill and mine site. A preliminary review of the pilot site sampling plans has begun.

Work Package 4 (WP-4): Implementation of the Procedures for Quality Assurance and Quality Control

The goal of WP-4 is to evaluate the current status and condition of the analytical capacity in the laboratories in the Member States responsible for analytical support of the monitoring programmes in the countries. This includes staff capabilities, experience, available equipment and required infrastructure as it relates to the measurement of specific radionuclides (uranium decay series) in a particular medium (air, soil or water). The purpose of such an evaluation is to identify missing elements and gaps in technical provisions of the participating laboratories and also to identify specific needs for further assistance. This WP is structured such that two ‘pilot’ analytical laboratories from each participating Member State will take part. In some countries, the number of laboratories willing to participate in the project activities may be extended in view of potential technical opportunities to carry out those or other types of analytical measurements within the framework of the integrated cooperative monitoring programmes. This evaluation, which is based on the results of a self assessment done by the national experts in their laboratories about their conditions and experience, has been completed through the use of a questionnaire and subsequent expert missions conducted in 2009. The IAEA’s Seibersdorf Laboratory has been involved in developing the WP and its implementation. The Siebersdorf Laboratory has designed, and the project will implement, a laboratory inter-comparison exercise (ICE). The first workshop on this ICE was held in Almaty, Kazakhstan in December 2009. The purpose of this workshop was to brief the project participants, to start preparing for the ICE and to provide them with a Quality Assurance and Quality Control (QA–QC) Guide. The exercise will consist of the analyses of uranium and thorium decay series radionuclides in samples of water and soils typical of what would be expected at areas affected by uranium production (mines and ore processing facilities). These samples are being prepared by the Siebersdorf Laboratory. The concentrations of uranium and thorium will be sent out as ‘unknowns’ to the participating laboratories for testing. The samples will be sent out in April of 2010.

The results of the exercise will be compiled in an IAEA report. This information will be utilized to identify individual laboratory strengths and weaknesses. Future WP activities will be adjusted accordingly. In preparation for the exercise, each of the laboratories participating in WP-4 has been provided with a set of reference samples with known radionuclide concentrations of uranium and thorium series radionuclides, the purpose of which is to enable participating laboratories regular self checking of the results of their analytical measurements.

Limited resources for the purchase of equipment have been provided to help the participating laboratories. This has included procurements for calibration sources, tracers, and some miscellaneous items needed for analytical procedures. Some training programmes were implemented in October and November 2009 in Kiev, Ukraine for the education of laboratory technicians in the use of certain analytical methods, QA–QC methodology and equipment.

There will be a second round of proficiency testing of laboratories near the end of the project in 2011 to measure progress.

Work package 5 (WP-5): Risk Communication and the Public

The primary goal of WP-5 is to assist operators and regulators in the Member States, who are responsible for developing and implementing the remediation programmes at the uranium production legacy sites on how to communicate risks and on methods for preparing and conveying public information. It is important to increase the awareness of the population living in the vicinity of uranium facilities about the hazards, giving them a better understanding of the ways to protect themselves from these potential hazards. WP-5 will include a workshop on risk and hazard communication for regulators and operators to be held in the fourth quarter of 2010. After the workshop, the Member States will be given the option of preparing a communication plan and of receiving IAEA support in the form of expert missions for the risk communication activities. Again a ‘pilot site’ and a community will be identified for this activity.

Work package 6 (WP-6): Institutional Controls as an Interim Risk Mitigation Measure

The single greatest risk reducing activity that the Member States of Central Asia could undertake is the implementation of institutional controls at the uranium production legacy sites. The purpose of WP-6 is to assist regulators and operators responsible for management and remediation of the uranium production legacy sites to establish optimal regulations and site institutional controls to prevent unauthorized entry to uranium legacy sites and unauthorized use of contaminated material from these sites. IAEA experts and consultants will work with Member State participants to evaluate specific situations at the pilot sites and will prepare specific proposals regarding institutional controls. These would be aimed at immediate near term risk reduction, long term guarantees of physical protection and safety of engineered barriers. In order to formulate optimal regulatory requirements for further use of the areas to be remediated, and to develop criteria for institutional controls at the legacy sites while planning the remedial action, during remediation and after remediation, long term stewardship is needed. This will be accomplished through a number of workshops and training courses for project participants involved with the pilot sites. The first workshop is planned for the third quarter of 2010.

Work package 7 (WP-7): Development of an Information Sharing Website

There is a well known sensitivity regarding cross border contamination for those Member States who share a common watershed and the drainage basin of the Syr-Darya river in Central Asia. In an attempt to build trust and confidence among the Central Asian neighbours, WP-7 will consist of the development of a website that will provide opportunities for information exchange among participants in the project, as well as between national and IAEA experts and project officers. The website will be developed by the IAEA in consultation with the Member States. The website will be hosted on the IAEA server to be accessible for use for project external and internal communication beginning in late 2010. The website will provide access on the open pages to be used for an exchange of information related to the project. Information such as reports, maps, figures, photographs, and videos will be available. The website will also have pages for restricted access on which information will be placed such as draft documents, monitoring programmes and data, instructions and guides for project activity applications. The goal will be to establish this communication and information platform and turn it over to the Member States at the end of the project. The information placed on the website will be provided by authorized web managers from the Member States. The authorization to place any information on the website will be under the control of the IAEA project technical officer and also the responsible website manager. The national project coordinator will control the information to be placed on the national web pages. It is intended to have the first phase of the website operational by the fourth quarter of 2010. The initial design is currently underway.

6. CONCLUSIONS

The international community has a strong interest in the environmentally and socially responsible systematic remediation of these uranium production legacy sites in accordance with international standards, recommendations and good practice. In order to accomplish this goal the following actions are necessary:

(a) Harmonization of national legislation and regulatory frameworks with the relevant international standards and recommendations;
(b) Preparation of environmental assessments prior to the commencement of remediation of the uranium legacy sites;
(c) Development of safety assessments to prioritize remedial actions;
(d) Development of remedial action plans;
(e) Implementation of remedial actions;
(f) Post-remediation monitoring and maintenance;
(g) Development and delivery of appropriate educational programmes for the regulatory bodies, mining and processing companies, relevant scientific institutions and representatives of the impacted communities;
(h) Development of national analytical capabilities.

Much progress has been made, yet there is still much more to do. The goal of the IAEA is to continue to actively contribute to the application of international safety standards and good practices as they relate to the remediation of uranium production legacy sites in Central Asia. The IAEA will continue to work with the Member States to build on the progress the Central Asian states have made since attaining independence while taking into account the common context in the region as well as the specific national context and requirements.

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Focus on Uranium: Invited Paper

EXTRACTION OF URANIUM FROM PHOSPHORIC ACID: GOOD PRACTICE

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Abstract

Prayon, a world leader in the phosphoric acid and phosphate salts industry, has good experience of uranium recovery from phosphoric acid: firstly, as a developer of the DEHPA/TOPO process, secondly, as an industrial producer of yellow cake through this process and, finally, as a designer of uranium extraction plants. This paper presents the history of the development of the DEHPA/TOPO process and gives a brief description of it. The main risks and some good practice to be adopted by potential investors are then outlined.

1. INTRODUCTION

For the phosphate industry, to talk again about uranium extraction from phosphoric acid means: **“Back to the future”**:

- “**Back**”, since the studies performed presently in the laboratories certainly prefigure the third generation of extraction units while those of the second generation have been stopped and in many cases have been dismantled for more than 12 years.
- **“To the future”**, for we are convinced of the relevance of this subject, in these times when society is questioning its development and the impact on the living environment and particularly on the global climate.

Sustainable development involves a rational management of worldwide resources. Therefore, it seems obvious that the ^{235}U resource, which is only used nowadays for producing energy, has to be recovered as much as possible and used as nuclear fuel in the power plants of the third or fourth generations, which are safer and more efficient, rather than remain diluted in phosphate fertilizers all over the farmlands of the world. Moreover, society would take more advantage of its oil resources if they were allocated as a priority to petrochemical activities rather than been turned into CO_2 within an energy production framework. Uranium recovery from phosphoric acid is a kind of project which fits perfectly with the 21st century in which ^{235}U will be the essential and inescapable fuel of a growing nuclear industry.

2. HISTORICAL BACKGROUND

As is often the case, it is interesting in this situation to recall where we are coming from in order to better understand where we are going. Solvent extraction of uranium from phosphoric acid saw its first industrial realization in Florida during the 1950s. However, this development was very fleeting because, in addition to the technical difficulties related to the instability of the solvents applied and to the accumulation of organic matter in the extractants, the uranium bearing concentrates obtained were so impure that they posed a problem during their treatment by the converters. At the end of the 1960s, a team from the Oak Ridge National Laboratory (United States of America), led by F.J. Hurst, developed a range of solvents generally made from neutral or acidic esters of phosphoric acid, with or without a synergetic agent such as phosphine. Thus, a range of processes, named according to the solvent used, was born — the DEHPA/TOPO, OPPA, MOPPA/DOPPA and OPAP processes.[1] Plants using one or another of these processes were built in the USA, Belgium, Canada, Iraq and Taiwan, China. The first to be operational was the Grace plant in Bartow, Florida (1976) and the last one to cease operation was the Prayon plant in Belgium (1998). In the meantime, the DEHPA/TOPO process had appeared to be the most efficient and some plants that began with another process subsequently converted to the DEHPA/TOPO process. Despite having a moderate power of extraction that required a two cycle process, the DEHPA/TOPO system revealed itself as the best one because of its good stability and selectivity. Indeed, at that time, the only reason to shut down the most highly performing DEHPA/TOPO units was economic: the uranium price dropped to less than 20 US $/kg. Despite some interesting work performed with new solvents in certain laboratories, the DEHPA/TOPO process in its latest evolution remains the reference process for any operator willing to avoid any industrial risk.

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[1] DEHPA/TOPO: di(2 ethylhexil) phosphoric acid/tri octyl phosphine oxide; OPPA: octyl pyrophosphoric acid; MOPPA/DOPPA: mono and di octyl phenyl phosphoric acid; OPAP: octyl phenyl phosphoric acid.

3. PRAYON AND URANIUM EXTRACTION FROM PHOSPHORIC ACID

At the end of the 1970s, the two Belgian companies Prayon and Union Minière, who were skilled in the phosphoric acid and non-ferrous metals industries, respectively, and who were both using liquid–liquid extraction technology, decided to share their expertise and joined forces to develop the industrial application of the DEHPA/TOPO process for uranium bearing phosphoric acid from Morocco. Based on pilot test results, two plants using mixer–settler technology were designed and built in Belgium.[2] With an annual capacity of 85 t of U_3O_8, those plants were operating from 1980 until 1998 and produced an average of 50 t U_3O_8 per year by treating all the available uranium bearing acid. During this period, many improvements were incorporated into the process, with the aim of improving the quality of the uranium concentrate produced, reducing the consumption ratios of additives and solvents and enhancing the reliability of the various items of equipment.

In 1981, in collaboration with the Belgian engineering company Mechim, Prayon became involved in an industrial project for uranium extraction in Iraq, working on every stage of the project: feasibility tests and sizing trials, equipment specification, drawing supervision, development of the operating manual, training of personnel in charge of operation and, finally, assistance to the startup that took place in 1984.

In 1998, because of the termination of industrial activity in Belgium for economic reasons, both plants, Puurs and Engis, were closed and dismantled. Before releasing the sites for further use, Prayon’s teams had to face the problematic decontamination of certain areas. For Prayon’s technicians, this decontamination phase, which was carried out under regulatory control, offered the opportunity to enrich our knowledge, since it challenged some preconceptions and the use of certain building materials and lining materials. A large number of the technicians involved in uranium extraction activities from 1978 to 1999, in the R&D Department, at the Central Laboratory, in the Engineering Department, as well as in the Production Department, are still working in the Prayon company today. This specific expertise, coupled with the worldwide recognized know-how of Prayon in the field of phosphoric acid production, is an indisputable and quite invaluable asset for any industrial company in charge of a project in the field of uranium recovery.

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[2] The first was in Puurs and the second was in Engis.

4. THE DEHPA/TOPO PROCESS

4.1. process description

The process consists of 4 steps or sections:

(i) The pretreatment of phosphoric acid;
(ii) The first cycle of extraction;
(iii) The second cycle of extraction;
(iv) Precipitation and conditioning of yellow cake.

A diagram of the process is shown in Fig. 1.

4.1.1. Phosphoric acid pretreatment

The pretreatment of phosphoric acid was jointly developed by Prayon and IMC at the end of the 1970s in the New Wales plant in Florida. From Prayon's point of view, the acid pretreatment is nearly as important as the extraction itself. Indeed, this step determines to a large extent the success of the solvent extraction processes that follow. High performance in the extraction step depends on the following:

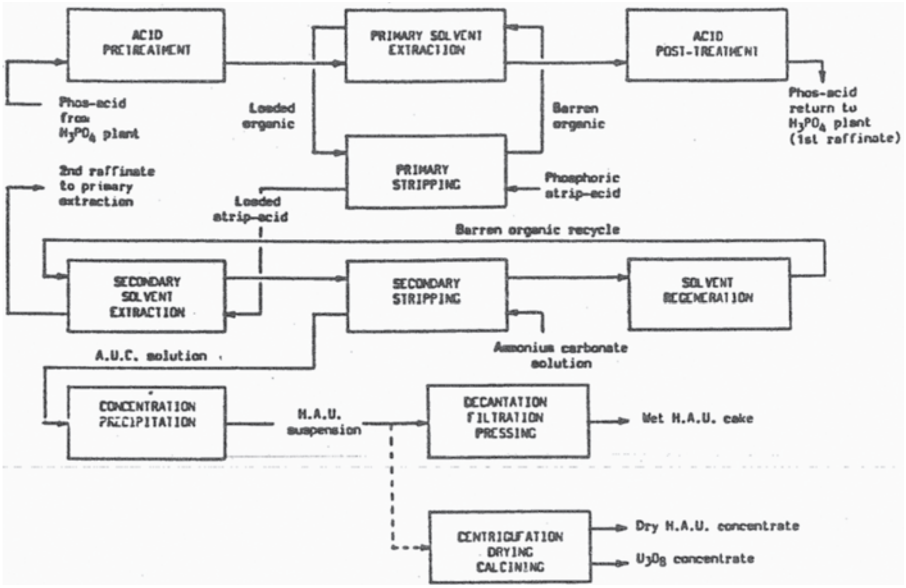

FIG. 1. The Prayon uranium recovery process.

(a) Strict control of the solvent and acid flows;
(b) A clear cut division between the two phases after each mixing;
(c) The absence of any ‘organic crud’ formation that would inevitably reduce the process efficiency and cause losses of acid and solvent.

For this reason, Prayon subjects the primary phosphoric acid to extensive preparation, including:

(a) Cooling;
(b) Desaturation;
(c) Clay treatment;
(d) Filtration;
(e) Absorption with activated carbon.

In these conditions, the resulting clear ‘green acid’ is completely desaturated, free of any mineral or organic solids and free of soluble humid material. It is perfectly prepared for further processing by liquid–liquid extraction.

4.1.2. First cycle of extraction

Both extraction and stripping (re-extraction) are carried out in a three or four stage battery of rectangular mixer–settler units. The DEHPA/TOPO solvent requires the uranium to be in the U^{6+} state for the extraction and, conversely, to be in the U^{4+} state for the reductive stripping. The judicious use of scrap iron added to a small flow of phosphoric strip acid to lower its redox potential during the stripping is a major factor for the correct operation of the plant. After the extraction step and separation of the entrained solvent, the main flow of phosphoric acid (the first raffinate) returns to the phosphoric acid production plant with the same clean aspect as the green acid.

4.1.3. Second cycle of extraction and refining

The same DEHPA/TOPO solvent is used in the second cycle but with slightly different concentrations. The strip acid from the first cycle enters the secondary extraction with a U_3O_8 concentration of 10–12 g/L. The secondary stripping is carried out with a solution of ammonium carbonate. It is performed within a precise range of conditions such that the ammonium–uranium carbonate formed (AUC or $UO_2CO_3 \cdot 2(NH_4)_2CO_3$) remains perfectly soluble, while other metallic compounds such as iron are precipitated.

4.1.4. Precipitation and conditioning

Uranium precipitates naturally by concentration of the clear AUC solution and gives a hydrated salt ($3UO_3{\cdot}NH_3{\cdot}5H_2O$) called HAU or yellow cake. The purity of this product lies well beyond the requirements of all converters. Two ways of conditioning are then possible:

(i) The HAU suspension is filtered and washed in a pressure filter and the wet yellow cake can be directly put in drums. This concentrate does not need any calcination and it does not create any problems concerning radioactive dust or environmental protection.
(ii) The HAU precipitate can be dried and, optionally, calcined. When calcined, the resulting product is a U_3O_8 concentrate with more than 80% U.

4.2. Site integration

During a project to extract uranium from phosphoric acid it is important to always keep in mind that the contemplated extraction unit will be adjacent to an existing phosphoric acid production plant. The new extraction unit will have to process an intermediate flow of process material. This involves constraints but also assets.

Constraints:

(a) The need for siting the first cycle of extraction close to the storage of phosphoric acid to be treated may lead to location or layout problems on an industrial site already largely occupied;
(b) The new activity of extracting uranium must not pose a threat to the realization of quantity and quality objectives or to the lifespan of the phosphoric acid production plant;
(c) The industrial culture among the supervisory and operating staff of a phosphoric acid plant is quite different from the one that has to be taught to the staff in charge of the new unit.

Assets:

(1) Some equipment of the phosphoric acid production plant may be allocated to the pretreatment section that precedes its admission to the first cycle of uranium extraction;

(2) The quality of phosphoric acid that comes out of the uranium extraction unit is greatly improved as a result of (i) the pretreatment, which comprises mainly desaturation (eliminating a certain quantity of impurities) and removal of organic matter and (ii) the subsequent removal of uranium;
(3) The workers allocated to the new extraction unit often have a very good knowledge of the main input, namely the phosphoric acid.

4.3. Prayon’s experience

4.3.1. New risks

Any phosphoric acid producer investing in a uranium extraction unit has to be fully aware of the new risks that have to be managed, namely exposure to intakes of radioactive material and to external radiation. Therefore, this new activity has to be done under the control of a ‘national approved organization’ that will ensure that the regulatory or legal measures are duly respected by the industrial operator. According to Prayon’s experience, this supervision occurs mainly at the second cycle, including the storage of the highly uranium bearing phosphoric acid coming from the first cycle of extraction (the ‘loaded strip acid’). During the industrial operation of this process by Prayon from 1980 to 1998, the workers having regular access to the second cycle of extraction were equipped with personal dosimeters deployed on a monthly basis and analysed by the national approved organization. Moreover, the same workers were given a specific annual medical examination in order to detect any potential intakes of radioactive material.

During the 18 years of operation and worker supervision, Prayon never had to redeploy a worker for reasons of internal or external radiation exposure. To achieve that result, it was essential to follow an integrated risk management approach from the outset, that is, during the design stage and the operational stage, the latter involving the implementation of strict work organization measures.

It is important to note that besides the risk linked to uranium itself, the risk linked to the use of an organic solvent has to be managed as well.

4.3.2. Design

The general design of the second cycle extraction unit must take into account the need for the following measures:

(a) Maintaining control over all outgoing flows and particularly to avoid any release of uranium into the environment, through:

- The provision of a containment wall for the storage tanks containing the loaded strip acid and second phosphoric acid raffinate, the tanks containing the acidic washing waters recycled to the fertilizer unit and tanks containing solvent — this containment, the volume of which must be sufficient to absorb the content of the biggest tank, is to be fitted with a sump that enables pumping out of any eventual leakage;
- The provision of a slightly sloping floor towards a gutter for that part of the plant in which extraction and subsequent operations take place (that is, where equipment containing phosphoric acid, solvent and slightly acidic washing waters is located) — thus any potential leak will be directed to an adequately dimensioned and fitted sump;
- The isolation and placing under a slight negative pressure of that part of the section where equipment contains yellow cake, that is, equipment for precipitation, filtration or centrifuging, drying and packing (vatting).

(b) Keeping all working zones free of any contamination, through:
  - The use of non-porous material for the construction or covering of walls and floors;
  - The avoidance of any locations that could be difficult to clean or to decontaminate during the finishing stages of the civil engineering work;
  - The covering of gutters with removable pieces made out of material that is resistant to acids and fuel and is easily cleanable;
  - The equipping of the section with a decontamination post for the treatment of small pieces and parts that need to be maintained externally and of objects to be disposed of such as used individual protection equipment including shoes, glasses, gloves and overshoes.

(c) Applying strict access control to the section, through:
  - The use of a security camera to monitor access to the control room;
  - The provision of two changing rooms for access and exit of workers and any visitors — a ‘green’ changing room where the operators keep their personal clothes and a ‘red’ changing room where working clothes and protection equipment are stored — between which are located a ‘humid’ locker room with showers and a ‘dry’ locker room, both equipped with a Geiger counter.

4.3.3. Work organization

Work organization must strictly limit the number of people authorized to enter the section, through:

(a) The establishment of operating teams comprising a process operator (a chemical engineering technician) and a maintenance operator (an electromechanical technician) who can internally repair small devices in a small workshop inside the section;

(b) A strict limitation on the number of external visitors, all of whom must wear a dosimeter, must be recorded in the visit book (name and dosimeter number) and must be checked at the exit with a Geiger counter.

4.3.4. Decommissioning and decontamination

If the industrial activity of extracting uranium is terminated, the site will have to be decontaminated before any possible dismantling is carried out and before the site can be released for further use. This decontamination will be easier if the principles mentioned above have been followed during the plant design phase. For economic reasons, Prayon had established its second cycle of extraction in a 19th century building. This was not a good choice. The walls, with a thickness of about 80 cm, were made up of extremely porous bricks. After decontaminating the equipment by washing with sulphuric acid, approximately 800 m^2 of wall surfaces were marked off in squares. Each square was numbered. The radioactivity of each square area had to be reduced to the background level. To reach this level, sometimes as much as 30 cm of the wall thickness had to be removed at certain locations.

Invited Paper

REVISION AND RECAST OF THE EURATOM BASIC SAFETY STANDARDS: STATUS REPORT AND PUBLIC CONSULTATION

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Abstract

The new ICRP Recommendations in Publication 103 introduce a distinction between three exposure situations (existing, planned and emergency exposure situations) and apply radiation protection principles to the different exposure situations. They have prompted a revision of the international standards and a revision of the Euratom Basic Safety Standards Directive together with a recast with other Euratom radiation protection directives. Although the present Euratom Basic Safety Standards address NORM industries as ‘work activities’, they do not prescribe how the protection of workers and members of the public should be regulated. This, as well as the identification of which industries were cause for concern, was left to Member States. Building materials were not explicitly addressed in the 1996 Euratom Directive, but the Group of Experts established under Article 31 of the Euratom Treaty established guidance for the control of radioactivity in building materials and defined a radioactivity index for this purpose. In another guidance report, the Group of Experts also defined exemption and clearance levels for materials processed in NORM industries and for residues from these industries. Building materials will now be included explicitly in the scope of the Euratom Basic Safety Standards and NORM industries will, if they are on the so-called ‘positive list’, be regulated as a planned exposure situation. The new Euratom Basic Safety Standards are close to a final draft and an outline of the requirements for natural radiation sources is presented. Within the process of revising the Euratom Basic Safety Standards, the European Commission launched in 2009 a public consultation on the new requirements for natural radiation sources. The outcome of the consultation is discussed.

1. RECAST OF THE EURATOM BASIC SAFETY STANDARDS

1.1. Recast

The European Commission (the Commission) is currently in the process of recasting five Euratom Council Directives: the Basic Safety Standards Directive [1], the Medical Directive [2], the Directive on High Activity Sealed Sources [3], the Directive on Outside Workers [4] and the Directive on Public Information [5]. A Council Directive is binding and Member States are obliged to transpose the requirements into national legislation. The aim of the recast is to merge the five Directives into one while keeping as much of the present texts as possible. The procedure does however leave room for certain parts to be revised and the Commission is taking this opportunity to clarify and strengthen legal requirements, based on the new ICRP Recommendations published in 2007 [6] and on experience gained by Member States and the Commission since the present Basic Safety Standards Directive (EU BSS) was adopted in 1996. The revision of the EU BSS coincides with the revision of the international BSS in which the Commission participates actively in order to achieve a high degree of harmonization between different international standards.

1.2. Exposure situations

The revision of the EU BSS will take account of the new ICRP Recommendations. The Commission has undertaken to structure the requirements along the concepts of planned, existing and emergency exposure situations, highlighting the role of optimization below suitable constraints and allowing for reference levels.

Even if the principles of protection according to the ICRP are very much the same for all exposure situations, the distinction between exposure situations matters in a regulatory context and precise definitions are needed. Unfortunately, the ICRP offers nothing but rather loose descriptive formulations. This introduces confusion between the concepts of a planned exposure situation and the planned operation of a new radiation source. While this approach is valid in the context of the application of the principle of justification, it is not the case when deciding on the appropriate mode and level of regulatory control. Some have concluded that “sources that already exist when a decision on control has to be taken” should always be managed as an existing exposure situation. The Commission strongly believes that when an activity significantly affects or alters an exposure pathway in a situation caused by existing sources, such as naturally occurring radioactive material (NORM) or cosmic radiation, this is a planned exposure situation. Hence NORM industries and the operation of aircraft or space craft are planned exposure situations and the activities can be labelled as practices. On the other hand, NORM with levels of activity concentration that are common in the earth’s crust should be exempted from the requirements for practices.

Existing exposure situations are in general those for which the exposure results from where you are, rather than what you do. In a dwelling, radon ingress from soil is not related to any human activity, so it yields an existing exposure situation. On the other hand, high levels of radon in the workplace are the responsibility of the employer and should be managed in the same way as occupational exposure in practices.

Another matter is the placing on the market of commodities, which clearly is a human activity. However, if the radioactive substances arise from an existing exposure situation, then it is more convenient to manage such commodities in the same context. Hence, building materials are managed as an existing exposure situation.

1.3. Natural radiation sources

1.3.1. Present BSS

Natural radiation sources are within the scope of both the present international standards [7] and of the present EU BSS. The international standards address radon at work (both ‘incidental to’ and ‘directly related to’ the work), but consider industries processing or involving NORM only as “any other practice specified by the regulatory authority”, and put more emphasis on discharges of effluents and disposal of radioactive waste than on exposure at work, such as external exposure or inhalation of dust. The European Directive introduced more specific radiation protection requirements with regard to natural radiation sources, for ‘work activities’ involving NORM (see Title VII of the Directive), for radon at work, and for the exposure of aircrew to cosmic radiation. The ‘work activities’ indicated in Title VII were specified as:

“a. work activities where workers and, where appropriate, members of the public are exposed to thoron or radon decay products or gamma radiation or any other exposure to radiation in workplaces such as spas, caves, mines underground workplaces and above ground workplaces in identified areas;
b. work activities involving operations with, and storage of, materials, not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of workers and, where appropriate, members of the public;
c. work activities which lead to the production of residues not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of members of the public and, where appropriate, workers;
d. aircraft operation.”

However, the Directive does not prescribe how the protection of workers and members of the public should be regulated. This, as well as the identification of which industries were cause for concern was left to Member States. Radon in dwellings was excluded from the scope of the Directive (but addressed in Commission Recommendation 90/143/Euratom [8]). Both the international and the European standards reflect ICRP Publication 60 [9] and in principle regard the exposure to natural radiation sources as an 'intervention' situation, even though the concept of 'work activities' in the European Standards is somehow in between practices and interventions.

1.3.2. Revision of the BSS

In 2005 and 2006, the Group of Experts established under article 31 of the Euratom Treaty set up a number of working parties to examine different parts of the regulatory framework. One of these working parties was dedicated to the management of natural radiation sources.

The Working Party (WP) Natural Sources worked in three steps, taking into account existing Commission Recommendations and earlier guidance. The first step was to look into present requirements for natural radiation sources, in particular NORM, and to suggest adjustments. The second step related to radon in dwellings, taking Commission Recommendation 90/143/Euratom into account. The third step was to set up new requirements for building materials containing naturally occurring radionuclides. As a result, the natural radiation sources that are explicitly incorporated in the draft EU BSS are:

(a) Naturally occurring radioactive materials used or processed in specific industries (NORM industries) or used as building materials;
(b) Indoor exposure to radon (^{222}Rn) in dwellings and workplaces;
(c) Exposure of aircrew and space crew to cosmic radiation.

This paper is essentially concerned with the new developments with regard to NORM industries and building materials, but for the sake of completeness, Section 5 is devoted to the management of radon exposure in existing exposure situations. It must be emphasised that radon exhaling from NORM in waterworks or in mines is managed in the same way as other exposure pathways, as part of a planned exposure situation. For the sake of completeness, it should also be mentioned that the existing requirements on the protection of aircrew have been kept without modification. The exposure of space crew will be managed as a specially authorized planned exposure.

2. REQUIREMENTS FOR NORM INDUSTRIES

2.1. Identifying exposure situations

The regulatory framework for NORM industries should essentially be the same as for other practices dealing with 'artificial' radionuclides. In the draft EU BSS, NORM industries have been integrated into the regulatory system as practices. However, as is shown further down, allowance is made for the fact that natural radiation sources are ubiquitous. It is also most unlikely that severe radiological accidents would occur and the existence of other industrial hygiene controls should be taken into account in the case of occupational exposure to NORM.

An important step towards harmonization of the regulatory framework for NORM is the introduction of a specific list of industrial activities of concern in the draft EU BSS:

(i) Extraction of rare earths from monazite;
(ii) Production of thorium compounds and manufacture of thorium-containing products;
(iii) Processing of niobium/tantalum ore;
(iv) Oil and gas production;
(v) Geothermal energy production;
(vi) TiO_2 pigment production;
(vii) Thermal phosphorus production;
(viii) Zircon and zirconium industry;
(ix) Production of phosphate fertilizers;
(x) Cement production, maintenance of clinker ovens;
(xi) Coal fired power plants, maintenance of boilers;
(xii) Phosphoric acid production;
(xiii) Primary iron production;
(xiv) Tin/lead/copper smelting;
(xv) Ground water treatment facilities;
(xvi) Mining of ores other than uranium ore.

This so-called 'positive list' covers industrial activities known to require regulatory consideration[1] and is to a large extent similar to the industry sectors

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[1] Uranium mining and uranium milling are part of the nuclear fuel cycle and require licensing.

listed by the IAEA [10]. Member States may add industrial activities to the list if the national authorities conclude that they deserve regulatory attention.

2.2. Materials of concern

The 1996 BSS Directive offered flexibility for Member States to take into account national circumstances for the identification of ‘work activities’ that would be of concern. Soon after the Directive was adopted, Member States brought forward the idea that there was merit in using the concepts of exemption and clearance as tools for the identification of NORM industries and for establishing the need for regulatory control. The Article 31 Group of Experts drew the following main conclusions:

(a) As a result of the large volumes of material processed and released by NORM industries, the concepts of exemption and clearance merge, and it is appropriate to lay down a single set of levels both for exemption and clearance;
(b) While the basic concept and criteria for exemption–clearance for ‘work activities’ are very similar to those for practices, it is not meaningful to define the levels on the basis of the individual dose criterion for practices (10 μSv per year); instead a dose increment, in addition to background exposure from natural radiation sources, of the order of 300 μSv is appropriate.

Similar to the approach for artificial radionuclides, on the basis of scenarios for public and occupational exposure, exemption–clearance levels for NORM have been calculated. The calculated values were rounded to 0.5 Bq/g for uranium and thorium in secular equilibrium (5 Bq/g for ^{40}K). Some individual elements in the decay chain, e.g. ^{210}Po or ^{210}Pb, warrant the use of significantly higher values, by up to two orders of magnitude. Numerical values can be found in the EC guidance Radiation Protection 122, part II [11].

For the sake of international harmonization it is now proposed that the draft EU BSS will introduce the same values for exemption and clearance as in the new international standards, while keeping the exemption criteria for natural radiation sources: annual doses of 1 mSv for workers and 0.3 mSv for members of the public. The values in the international standards will be based on earlier work leading to IAEA Safety Guide RS-G-1.7 [12]. For radionuclides of natural origin, this work was based on the concept of ‘amenability to control’, underlying the concept of exclusion. Values were eventually chosen at the upper end of the distribution of concentrations in soils around the world (UNSCEAR data [13]): 1 Bq/g for radionuclides in the U and Th decay series and 10 Bq/g for ^{40}K, see

Table 1. This is a factor of two higher than the values derived in RP122, Part II. The difference of a factor of two may be viewed as small, but is still causing a lot of concern. Although the ICRP in Publication 75 [14] confirms that workers or members of the public will generally not receive doses higher than 1 or 0.3 mSv per year, respectively, if the activity concentrations are lower than the values in Table 1, this is not always the case for building materials. These are now regulated in their own right in the draft EU BSS and special consideration is given to NORM industries on the ‘positive list’ producing residues used in building materials, see Sections 3 and 4.3.

2.3. A graded approach to occupational exposure

For industries on the so-called ‘positive list’, the regulatory authorities should be notified by the undertaking if, at any point in the industrial process, the activity concentration in any material exceeds the values indicated in Table 1. The notification should include information on the materials processed, radionuclide concentrations, products, by-products and residues. Based on this information the authorities will decide on the level of regulatory control and can impose requirements for the initial assessment of the exposure of workers. When the assessed exposure of workers is expected to be less than 1 mSv per year, the practice may be exempted from further regulatory control. If the assessed exposure of workers in a group of identical industrial processes is consistently less than 1 mSv per year, these processes could be exempted on a generic basis.

TABLE 1. VALUES FOR RADIONUCLIDES OF NATURAL ORIGIN IN EQUILIBRIUM IN SOLID MATERIALS[2]

	Activity concentration (Bq/g)
Radionuclides from the ^{238}U series	1
Radionuclides from the ^{232}Th series	1
^{40}K	10

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[2] The values in Table 1 are those recommended in IAEA Safety Standards Series No. RS-G-1.7. They were derived using the concept of exclusion and are based essentially on the distribution of concentrations in the earth’s crust.

If the exposure of workers can exceed 1 mSv per year but is less than 6 mSv per year, Member States must consider authorization, that is, either registration or licensing, or consider whether circumstances indicate that the practice should be exempted from further regulation. Member States must require the employers to regularly assess whether doses could effectively be further reduced and whether there is a potential for doses to increase over time or as a result of changes in the work practice. Where appropriate, the authorities must request a formal implementation of the principle of optimization as it would for any other practice. Furthermore, designated members of the staff should be trained in radiation protection. For this purpose the competence and human resources of a health and safety department within the undertaking can be used. Where appropriate, a radiation protection expert should be responsible for the training and for further advice on protective measures.

When the exposure of workers exceeds or is likely to exceed 6 mSv per year in normal operation, Member States must require that the practice be licensed.

3. REQUIREMENTS FOR BUILDING MATERIALS

The present EU BSS do not include specific requirements related to radionuclide concentrations in building materials. By introducing such requirements in the draft EU BSS, the Commission on the one hand complements the Council Directive on construction products [15], on the other pursues further harmonization of the regulatory approaches by Member States to allow free movement of building materials within the European Union.

3.1. List of building materials and components

Member States will be required to insert into their legislation a list of the different types of building materials which need to be controlled with regard to their emitted gamma radiation. When setting up this list, an indicative list in an Annex to the BSS must be taken into account. The list of materials will contain:

(a) Natural materials:
  - Alum-shale;
  - Building materials or additives from natural igneous origin, such as granite, gneiss, porphyries, syenite, basalt, tuff, pozzolana and lava;

(b) Materials incorporating residues from NORM industries such as flyash, phosphogypsum, phosphorus slag, tin slag, copper slag, red mud (residue from aluminium production) and residues from steel production.

The requirements will offer the possibility for Member States to add or withdraw certain categories of materials when there is evidence of them being a cause for concern or not being of concern.

3.2. Measurements and activity concentration index

For the materials of concern, the industries placing such materials on the market will be required to measure the three radionuclides indicated in an Annex. The activity concentration index (I), as defined in EC guidance Radiation Protection 112 [16], has to be calculated.[3] The industry is required to provide information to the national authority on the results of the measurements and of the corresponding activity concentration index (I).

3.3. Reference level and classification

A reference level of 1 mSv per year is proposed for indoor external exposure from building materials. This is in excess of the background outdoor external exposure. When assessing compliance with the reference level, the doses from the exposure to local prevailing activity concentrations in the undisturbed earth's crust should be subtracted.

If the building material is found to give doses not exceeding the reference level, then the material should be exempted from requirements, except for appropriate further monitoring of activity concentrations if so required. If the material is liable to give doses exceeding the reference level, the authority should consider appropriate control measures ranging from registration and general application of relevant building codes, to specific restrictions on the use of such material. Building materials not exceeding the reference level would be exempted from any restrictions on the market in the European Union.

Two groups of building materials should be considered:

(i) Materials used in bulk amounts;
(ii) Superficial and other materials with restricted use.

For this purpose the activity concentration index I could be used for the classification of the materials into four classes leading to two categories of building materials (A and B), as shown in Table 2. The distinction of materials

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[3] $I = C_{Ra226}/300\ \text{Bq/kg} + C_{Th232}/200\ \text{Bq/kg} + C_{K40}/3000\ \text{Bq/kg}$, where C_{Ra226}, C_{Th232} and C_{K40} are the activity concentrations in Bq/kg of the corresponding radionuclides in the building material. For practical purposes the measurement of ^{232}Th can be replaced by that of ^{228}Ra which is more readily measured.

TABLE 2. VALUES FOR THE ACTIVITY CONCENTRATION INDEX (I) IN BUILDING MATERIAL

	Category (corresponding default dose)	
	A (≤1 mSv)	B (>1 mSv)
(1) Material used in bulk amounts	A1 $I \leq 1$	B1 $I > 1$
(2) Superficial and other material with restricted use	A2 $I \leq 6$	B2 $I > 6$

into (1) or (2) should be defined by the national authority and based on national building codes.

The information relevant to the implementation of building codes should be made available before the materials are placed on the market. Relevant information includes radionuclide concentrations, activity concentration index and corresponding classification.

4. REQUIREMENTS FOR NORM EFFLUENTS AND RESIDUES

4.1. Effluent

As explained in Section 2.2, the draft EU BSS incorporates the activity concentration values recommended in IAEA RS-G-1.7 (see Table 1), but also keeps the exemption criteria for natural radiation sources: annual doses of 1 mSv for workers and 0.3 mSv for members of the public (to be compared with 10 µSv for artificial radionuclides). The criterion of 0.3 mSv per year applies to industrial facilities discharging liquid and gaseous effluents containing NORM. In general the criterion is satisfied if activity concentrations do not exceed those in Table 1. Guidance on the establishment of discharge authorizations in terms of total annual activity has been offered in Radiation Protection 135 [17].

4.2. Waste disposal

Only in exceptional cases (for instance, pipeline scale in the oil industry) do NORM industries give rise to residues that qualify for disposal in specific radioactive waste repositories. In most cases, residues from NORM industries will be disposed of in industrial landfill sites, provided they comply with the criteria for clearance. It should be emphasised that the values laid down in Table 1 do not allow for leaching and possible contamination of drinking water as an exposure pathway. The generic, conservative, inclusion of a drinking water pathway in the assessment of doses to members of the public from landfill sites could lead to very low activity concentration values. It is thus necessary to monitor drinking water for such possible contamination and for compliance with drinking water standards (as required under Council Directive 98/83/EC [18]). In any situation where there is an indication that such contamination may arise, a NORM industry may be subject to authorization even if the concentration values in Table 1 are complied with.

4.3. Recycling of NORM residues in building materials

The dose criterion of 0.3 mSv per year also applies to industries producing residues that are known to be recycled in building materials. Reuse and recycling of residues from any authorized practice is subject to authorization. The clearance of residues for the manufacture of building materials may warrant lower values than those proposed in Table 1. The building materials should be tested against the index value in Table 2. The reference level of 1 mSv applies to the building material as a whole, while the clearance criterion of 0.3 mSv applies only to the dose resulting from the incorporation of specific residues. Industries known to produce residues liable to cause the index for building materials to exceed 1 have to be notified to the regulatory authority. The authority may impose specific licensing conditions on such industries or allow the recycling of such residues if this is justified. They may also allow mixing of residues with other materials in order to reduce the value of the index if this is justified.

5. REQUIREMENTS RELATED TO RADON EXPOSURE

5.1. National action plan

The Commission Recommendation from 1990 [8] addressed the protection of the public against indoor exposure to radon, but exposures to radon in dwellings were excluded from the 1996 BSS Directive, while a flexible approach on radon in workplaces was adopted in that Directive. The Commission now proposes to strengthen this legal framework with requirements on Member States to enhance their national strategies for reducing radon exposures in dwellings and workplaces. One of the main requirements in the draft EU BSS is the demand for an action plan on how to manage long term radon exposure both in dwellings and in workplaces, for any source of radon, including soil, building materials and drinking water. The action plan should cover the issues specified in an Annex to the BSS, which include:

(a) Goals and targets, in terms of reduction of lung cancer risk;
(b) Criteria and available data used for the delimitation of radon prone areas;
(c) The basis for choosing and establishing reference levels in accordance with the EU BSS;
(d) Assignment of responsibilities, and coordination of national authorities and resources;
(e) Strategies, methods and tools for measuring radon levels, and for taking remedial actions, particularly in dwellings;
(f) Audits and reviews of the implementation of the plan;
(g) A strategy for communication to increase public and local decision makers' awareness of the risks of radon and its relation to smoking.

5.2. National reference levels

It is the responsibility of national authorities to establish national reference levels, taking into account the prevailing economic and societal circumstances, and then to apply the process of optimization of protection in their country. On the basis of the link between exposure to radon and lung cancer established by pooled analyses of residential case control studies [19–22] and the convergence of international recommendations from WHO, ICRP and IAEA[4] on the national strategies to be set for radon in both dwellings and workplaces, the Commission now proposes that Member States establish national reference levels not exceeding (as an annual average):

- 200 Bq/m^3 for new buildings;
- 300 Bq/m^3 for existing dwellings;
- 1000 Bq/m^3 for existing workplaces and other public buildings.

The Commission also proposes that if a workplace, despite actions carried out to reduce radon concentrations in line with the principle of optimization, still exceeds the national reference level, then the relevant and appropriate requirements for occupational exposure shall apply.

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[4] Reference is made to the WHO Handbook on Radon (2009), the ICRP Statement made in Porto, November 2009, and an IAEA technical meeting in Vienna, December 2009.

6. PUBLIC CONSULTATION

‘Natural radiation sources’ is one of the areas where the requirements in the new EU BSS will be more explicit compared with the requirements in the current BSS Directive. This, in combination with the Commission’s policy for enhanced transparency, prompted a public consultation on the proposal with regard to natural radiation sources. The consultation involved the organizations, authorities and industries affected by the Directive in order to achieve a better understanding and acceptance of the new ideas. It will also benefit the preparation of the impact assessment which accompanies any legal proposal in the European Union.

A consultation document with the Commission Services’ considerations regarding natural radiation sources (NORM, radon and building materials) in the new EU BSS was published on the Commission's website in February 2009. The consultation period continued until end of April 2009.

6.1. Contributions received

In total, 45 contributions were received, mostly from industry organizations (around 15) or governmental organizations or authorities (around 15). A substantial amount of contributions came from individuals (10) and from radiation protection associations or groups of experts (5). The contributions from industry came from the following sectors:

(a) The steel industry;
(b) The zirconium chemicals producers;
(c) The producers of abrasive products;
(d) The titanium dioxide producers;
(e) The building materials industry, including the tiles and bricks industry;
(f) The nuclear industry (FORATOM–ENISS).

With regard to the geographical distribution, the vast majority came from Member States within the European Union, mostly ‘old’ Member States. The consultation was well received and a large part of the contributors expressed their appreciation for being invited to comment on ideas this early in the process of revising the Directive.

In general the contributors endorse the goal of the Commission to harmonize, clarify and strengthen the requirements related to natural sources. They also believe that the Commission has chosen the right approach when introducing the so-called graded approach to regulatory control but they would like to have more information on the regime of notification, registration and licensing. There is a high demand for guidance and clarification about the rationale for certain issues and about how to implement the requirements in practice. Examples of such issues are:

(a) Why NORM industries are considered as planned exposure situations when ICRP Publication 103 considers them as existing exposure situations;
(b) The background to the values chosen as the upper boundary for radon reference levels;
(c) Why the exemption level for NORM industries with regard to doses to the public corresponds to 0.3 mSv per year whereas the level for building materials corresponds to 1 mSv per year and the upper boundary for radon in dwellings corresponds to several millisieverts per year.

Furthermore there is a demand for clear definitions of terms such as building, dwelling, reuse, recycling, disposal, waste, construction, natural radiation source and inert material.

6.2. Result of consultation

The comments received have been very useful in the further discussions and they have led to an improvement of the text in the Directive. For example:

(a) Terminology has been discussed and definitions have been introduced or modified;
(b) The so-called ‘positive list’ has received two additional sectors which are geothermal energy production and mining of ores other than uranium ores;
(c) The situation when NORM industries may affect drinking water pathways is now addressed explicitly;
(d) The list of building materials of concern has been modified according to suggestions received in the consultation;
(e) With regard to radon in workplaces, the suggested threshold for recording exposure to workers has been abandoned and the proposed upper boundaries for reference levels now mention that the working time (that is, the actual time in a radon environment) is an element which can be taken into account for workplaces.

The Commission is planning to further elaborate on principal issues and on the implementation of the new requirements in a guidance document which would be published in connection with the adoption of the new Directive.

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Invited Paper

A COMPETENCY SPECIFIC APPROACH TO EDUCATION AND TRAINING NEEDS IN NORM INDUSTRIES

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Abstract

Industries working with or processing NORM recognize an urgent demand for formalized education, training and professional development. MiLoRAD marks the first step in a systematic attempt to codify and teach in a coherent and consistent way the knowledge and skills that are required for all industries working with NORM. The apprentice based, company or even site specific model of training will continue to have its place; but the visibility and economic significance of industries working with NORM make it essential, not least for stakeholder confidence, to address their training and professional development needs in a more formal, transparent, competency based manner. A solution is urgent given the rapid development of demand for NORM industry manufacturing capabilities in emerging and developing economies, some with little or no indigenous source of knowledge or experience in the domain as they are manufacturing and marketing NORM-impacted products for the first time. The paper sets out the five tier, criterion referenced competency and best practice model determining the shape of MiLoRAD. It positions it within the context of HR needs in the phosphate industry in particular, as a first test case, underpinning a management and operational culture focused on safety, sustainability and efficiency. It proposes adherence to global competency standards, and, in association with suitable national or regional centres of excellence, explores options for certification or accreditation of both individuals and operating units who have successfully completed the courses. Training, CPD and wider educational needs are addressed in a combination of in situ (in post) learning, classroom and web based course elements. The applicability of the model to other NORM industries is also explored.

1. INTRODUCTION: NORM INDUSTRIES OR INDUSTRIES WITH ASSOCIATED NORM

The challenge of regulating industries using or processing naturally occurring radioactive materials (NORM) is still in its relative infancy. At an existential level, it is unclear how best to approach the issue given that NORM occurs naturally, is ubiquitous and presents a largely unavoidable, if very low level, risk. In this context, there is a valid case for asking whether NORM should be regulated at all. If the case is then made that regulation should be applied, or at least considered, from the moment NORM engages with technology and commerce, the challenge then focuses on defining what the purposes of such regulation should be, and how to measure success. Assuming that in general terms, the purpose will be to protect occupational, public and environmental health and safety, it is clear that NORM specific training and education programmes must address, and deliver, measurable competency in both process and outcome. Over time, the validity of the competency model used will be tested and refined through a combination of best practice (demonstrably safer process) and of accumulated evidence (demonstrably beneficial outcome). The combination may even become so compelling that it warrants the implementation of an enforceable standard to which industry is required by law to adhere.

While process may be largely circumscribed within the world of industry and commerce, in terms of outcome, any NORM industry, along with all industries in the age of sustainability, now has to account for itself in terms of stakeholder interest and benefit as well as financial return to shareholders. The emerging standard for analysing such accountability is the so called ‘triple bottom line’ (TBL) method [1] which submits all industrial activity to analysis according to its financial, social and environmental return. The TBL outcome measurement method is followed in this paper, with significant, but at least predictable, consequences for the remit of training and competency building for the NORM workforce.

It is much harder to propose a unified approach to training because there is no coherent, intrinsic definition of what a NORM industry is. In the absence of such a definition, there is a risk that a loosely framed label be applied to a highly heterogeneous set of activities whose only common feature is risk of exposure to naturally occurring radionuclides. It is already evident in the regulation of some NORM industrial activity, such as processing phosphate rock for fertilizer, that such an approach can have unwarranted and costly outcomes both to industry and to stakeholders.

1.1. What is a NORM industry?

While grouping various industrial activities together under the rubric 'NORM industry' may be convenient, it is a significant departure from conventional classification practice. Industry classification systems, such as the Standard Industry Classification (SIC) or the North American Industry Classification System (NAICS), or the more recent Industry Classification Benchmark (ICB) [2], are based on a taxonomic (rule based) model that takes defining operational characteristics of those industries and normalizes them to up to four levels of attribute. These attributes are summarized as a four digit code. For example, the ICB paper industry (code 1737) is part of the sector Forestry and Paper (code 1730) which in turn is classified as one of the Basic Resource industries (1700) which is aligned within the class Basic Materials (1000). No taxonomy is perfect, but at least the classificatory rules are apparent. This classification process allows for consistent and meaningful comparisons to be drawn between members of a given sector for benchmarking or performance analysis including operational indicators as a whole and overall health and safety outcomes in particular.

If, as seems to be the trend, NORM industries are to be grouped together for regulatory purposes [3], meaning that common standards and outcomes measures are to be applied to them, then those industries might reasonably expect that there is an operational basis or rationale for such a grouping. But in practice this is not the case. What actually happens is that a number of industries with one circumstantial attribute in common — dealing with NORM — are listed as if they belonged to a coherent, discrete class. For example, according to the IAEA [4, 5], NORM industries include:

(a) The mining and processing of **uranium** ores;
(b) Extraction of **rare earth** elements;
(c) Production and use of **thorium** and its compounds;
(d) Production of **niobium and ferro-niobium**;
(e) **Mining** of ores other than uranium ore;

(f) Production of **oil and gas**;
(g) Manufacture of **titanium dioxide** pigments;
(h) The **phosphate** industry;
(i) The **zircon** and zirconia industries;
(j) Production of **tin, copper, aluminium, zinc, lead, and iron and steel**;
(k) Combustion of **coal**;
(l) **Water** treatment.

The list has no taxonomic coherence; selection criteria include (a) whole industries, such as oil and gas, phosphate and zircon, (b) particular processes, such as manufacture of titanium dioxide pigments, (c) generic processes affecting most manufacturing industries, such as water treatment, and (d) even one category defined by not being another: ‘mining of ores other than uranium ore’. While it may be argued that in a world dominated by stakeholders with a high degree of sensitivity to anything to do with ‘radioactivity’, the common factor or attribute of using or processing NORM may be a necessary condition for grouping them together, it cannot be argued that the mere act of listing industries under a convenient administrative heading NORM is a sufficient case for treating them as a coherent group which deserves to be overseen and regulated as a single entity in a single way.

This is equivalent to describing all industries that manufacture products that might have toxic effects if ingested as ‘toxic’ industries. As Paracelsus recognized: “Alle Ding' sind Gift, und nichts ohn' Gift; allein die Dosis macht, daß ein Ding kein Gift ist” [Everything is poison, and nothing is poison free; it is only the dose that determines if a thing is not poisonous]. The challenge is to define under what circumstances, or at what points, if any, might radionuclide activity concentrations reach such a level that there is actual risk to humans or the environment from them.

1.2. Safety — an evolving concept

Just as a coherent definition of a ‘NORM industry’ is an elusive concept, so too is safety, which in an industrial context, at least as an outcome measure, is undergoing significant change as concepts such as sustainability and TBL return exert their influence. The consequence is that “Systematic qualification training programmes are required”, from which a culture of safety is a necessary and inevitable outcome, rather than something that is designed in [6]. The culture of safety does not set aside previous objectives, measures and targets. Physical and mechanical measures protective of life and limb, such as came in during the nineteenth and early twentieth centuries, remain as necessary as ever. Equally, objectives focused on operational efficiency remain current, with their associated metrics such as lost time injuries (LTI) per million hours worked. But the explosion of the BP refinery in Texas (2005), resulting in 15 fatalities and 180 injured, was a strong reminder that safety cannot be measured simply by the LTI rate [6].

Based on this and our own comparative analysis, the approach taken in this paper is that there are no intrinsic factors that justify grouping industries as NORM from an operational or process point of view — each so called NORM industry is different. But there is one extrinsic factor on which a common approach to training requirements can be based, the notion that each may be analysed according to the hazard analysis critical control point (HACCP) methodology [7], yielding in each case a limited but significant number of critical control points (CCPs) or safety barriers at which hazards, real or perceived, may be monitored, mitigated or, best, prevented. For each CCP a so called training and safety ‘hurdle’ or layer of protection (LOP) [6] may be put in place, of which the last hurdle, competency, is focused on the CCP always most prone to failure, the human operator. The training methodology then aligns the response options available to individuals and organizations dealing with such risks and hazards at each hurdle, and teaches best, safe practices according to the severity of the inherent risk and the competency of the individual exposed. The result is a training regime such as MiLoRAD, based on validated, continuously updated process safety knowledge.

1.3. NORM specific education and training requirements — focus on competency

For the negative reason that NORM industries are not amenable to assimilation into a single class, but for the positive reason that a competency based approach offers the best prospect of meeting the diverse operational requirements they face, we have adopted a highly disaggregated, competency based approach both to training needs analysis and delivery. Each trainee or trainee group is profiled by requisite competency, using competency first as a diagnostic tool to identify deficiencies in knowledge, skill and expertise and then as a goal. The gap between current and proposed future competency allows the creation of a tailored training or education programme suited to the individual or working group, a process that can also factor in other requirements such as working culture, language and wider stakeholder influences and expectations. This in turn creates a robust platform for capacity-building.

2. A COMPETENCY BASED APPROACH

Competency based training (CBT) is a systematic, knowledge and skills based approach to vocational education and training that focuses on what a person can do in the workplace as a result of completing a programme of job or task specific training. A competency may be defined in terms of what a person is required to do (operational task), under what conditions it is to be done (operating conditions), what the task is intended to achieve (outputs and outcomes) and how well it is to be done (performance standards). Competencies commonly map to skills, which in turn may be simple or complex in nature.

We propose that skills be broken out into at least five distinct categories:

(a) **Task specific** — such as using the clutch on a motor car or fitting a plug to a wire;
(b) **Procedural** — in which tasks are concatenated into a coherent process or flow sheet;
(c) **Contextual** — such as demonstrating capacity to work within the wider operating or process environment;
(d) **Contingent** — such as dealing successfully with the unexpected or unforeseen;
(e) **Interpersonal** — such as communications (oral and written) and teamwork.

In some cases, such as medicine, the job in question cannot be awarded at all unless the applicant is able to demonstrate competence in the relevant field. In some cases, this training is carried out as part of a compliance programme based on the need for each employee to meet the competency requirements set out in a company's standard operating procedures (SOPs). In the case of new employees, it will not be expected that they can meet competency requirements before they are hired; but they must meet these requirements to continue in post. In some industries, such as pharmaceutical, regulators and inspectors have the powers to visit sites and check compliance with SOPs, a process that often starts with inspection of training records. Most SOP driven regimes require not just that training be given to new employees but that all employees submit to regular refresher and update courses, typically every two to three years. These courses form part of the records that both the individual and the organization must keep to show compliance with the overall aims and objectives of the safety and quality standards within which competency based training for the most part sits.

Competencies may be broken out into different categories, such as 'essential' and 'universal' or 'global'. An essential competency is one that is so critical for a particular job that that job cannot be performed without it. A ‘universal’ competency is one that is required of all members of staff in an organization, regardless of job title. An example of a universal competency might be understanding of and compliance with the organization’s mission.

Competencies may be gained in many different ways, such as work and life experience, formal or academic education, apprenticeship, self taught programmes and job specific training and development programmes. Managers and employees work together to achieve a clear and consistent understanding of what level and type of competency is required to meet organizational performance requirements. Assessment of job performance over time determines the ‘overall employee competence’ level, both individually and collectively. It can be the case in engineering and manufacturing environments that too much emphasis is placed on individual and mechanical skills, at the expense of teamwork and ‘soft’ skills, such as communications.

Competency based training programmes often comprise modules, which may be further broken out into segments; each module segment leads to a so called learning outcome, such as a skill or learned procedure. Outcomes are based on standards set by experts, usually working in a coordinated fashion, drawn from industry, academia and government. Where a body of knowledge and experience is of strategic significance for a whole industry it is common for that industry to support, or work closely with, a centre of excellence (COE). In such cases, not least to preserve its independence, that COE will draw its funding, in preferably equal measure, from its constituencies, with no single stakeholder having the power to unduly bias the work that the COE undertakes.

Assessment is a continuous process that runs in parallel with learning, designed to ensure each student has achieved all the outcomes (skills and knowledge) required by each module. An initial diagnostic assessment is used to determine the initial level of competency of the learner; this may be conducted through a ‘pre-test’ in the form of a questionnaire, or an oral interview, combined with self assessment. A final assessment is typically conducted in a combination of ways including multiple choice questions, in situ demonstration of capability (for example, by the successful execution under examination conditions of job-critical tasks, in compliance with SOPs) and varying combinations of written and oral examination, often by a panel of experts. Using the device of a pre-test before an intensive programme and a post-test conducted after it is one way of measuring the impact of the training, for example in the scores obtained by participants before and after the course. But such techniques in isolation can be misleading since organizational performance is more dependent on what is retained in the longer term, something that can only be measured through time.

Ideally, progress within a competency based training programme is not based on a time based curriculum. As soon as students have achieved or demonstrated the outcomes required in a module, they can move to the next module. In this way, students may be able to complete a programme of study much faster. This has significant potential benefit for organizations with a high labour turnover or where particular tasks and skill sets critical to its operation and performance are held by one or a small number of individuals whose absence or unexpected departure can render the organization very vulnerable. But it is also at odds with current orthodoxy in many continuous professional development programmes, through which many professionals retain their professional qualifications; for such programmes are typically based on gaining ‘credits’ based on hours of training attended, many of which have no assessment component at all. These in our opinion are of questionable value.

3. THE COMPETENCY PYRAMID — APPLYING A CLASSICAL FIVE TIER COMPETENCY MODEL

One of the most seminal competency models was that developed in response to the 1980s pursuit of machine intelligence [8]. This broke competency out into a five tiered, progressive learning model as follows:

(a) Novice;
(b) Advanced beginner;
(c) Competent;
(d) Proficient;
(e) Expert.

The training needs of each level are addressed in a modular fashion, as described in Section 2, the system being ‘learner centred’ or ‘learner driven’, meaning that unlike academic institutions which hold examinations only at specific times of the year, the trainee or learner moves at the pace dictated by their own understanding and competence. So the learner can move up the pyramid in discrete steps according both to personal drive and ambition and the needs of the employing organization or company.

The model is generic and does not assume a particular body of knowledge or experience as a starting point for training, but rather an outcome or end point. It is also adaptable in that it not only recognizes that there are widely differing, highly specific processes to which the competencies envisaged may be applied, but also the industrial settings in which they are required may be at very different points in the product life cycle. In the early stage (new product development), the competencies themselves may be an object of study and research, while in a mature market situation with a reference product, training may be at a minimum based on best practice, or even embodied in the requirements of a standard.

That said, at the level of hurdle theory, or the use of required ‘layers of protection’, there are certain competencies that are non-negotiable in nature that all employees need to understand and practice to a certain level of proficiency, or risk exclusion from the site or even employment. These are akin to the emergency stop procedures in driving tests — you will fail the driving test on failing that one competency, however good the other skills you demonstrate may be. It is not yet clear if NORM may require such a non-negotiable skill set. It would clearly be essential to have one in place if the evidence base indicates this should be so. But to impose one simply because it is felt there should be one is misguided.

As shown in Fig. 1, the learner may progress up the competency pyramid in discrete steps, each step being associated with a change or increment in the nature and degree of responsibility shared for the safe, efficient conduct of a NORM related process. This progress maps to a competency state, to a market status and to a level of acceptance in terms of validation and degree of enforceability, from justification in terms of sheer expertise as a foundation, to justification as a legal requirement as a probable but not essential end point.

3.1. Tacit knowledge — the paradox of the inarticulate expert

A paradox addressed, but not resolved in this model, is the concept of ‘tacit knowledge’ in its description of the highest level of competency, the expert. As

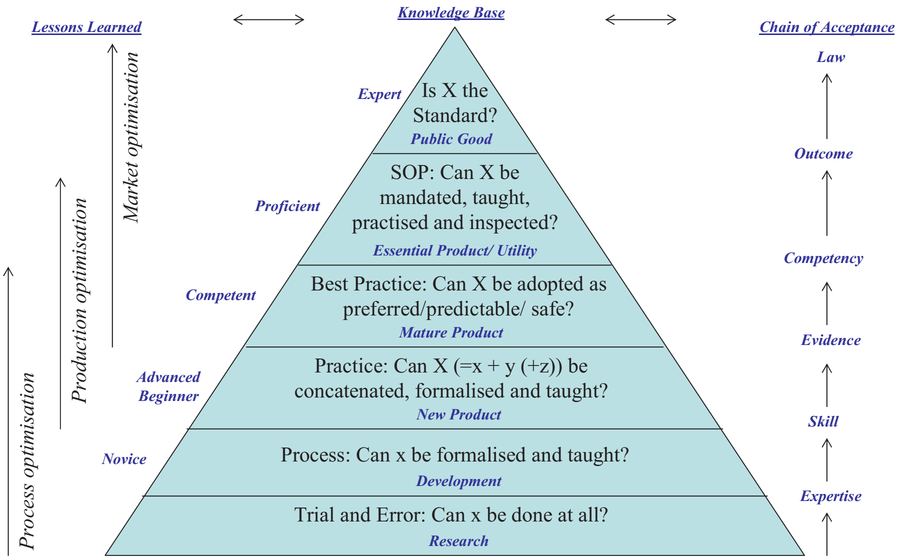

FIG. 1. *The competency pyramid: from trial and error to standards?*

explained in Refs [9, 10], experts (level 5) can appear to know less about a domain in which they are expert than those who are merely competent (level 3) because their knowledge is so deeply compiled and intuitive they have difficulty in explaining it, often leaving major steps out in their reasoning or in their explanations of solutions. The competent person by contrast will list out every detail, as may be demanded by the culture of SOPs. The paradox, therefore, is that while the experts may be the best or only source, or validator, of competencies, and hence of SOPs, they may not be the best person to draft them. Not by coincidence the MiLoRAD system referenced in this paper was built by a team chosen in part because of its blend of different levels of competence, in this case in NORM and health physics as well as the practices and processes of the phosphate industry.

3.2. The competency matrix

Table 1 sets out the underlying competency matrix on which MiLoRAD is based, using the phosphate industry as a worked example:

It is clear that competency based approaches to radiation safety training in general are starting to attract attention at government level [11]. Following the maxim that it is generally better in regulatory matters for an industry to lead than be led, the NORM sector may wish to embrace the model purposefully and without delay.

4. DELIVERY: A GLOBAL NETWORK OF CENTRES OF EXCELLENCE

To facilitate delivery to the phosphate sector in particular, an international network of centres of excellence working on various aspects of the phosphate value chain has been formed, with the Florida Institute of Phosphate Research as its coordinating hub. Counterparts in a number of countries including Brazil, Jordan, Morocco, and Spain are in place. Certification programmes based on the competency model are well advanced and will be available to interested parties from 2010.

TABLE 1. COMPETENCY MATRIX

Level	Generic	NORM operational	(Radiation) safety hurdle	Management
1. Novice	• Works to taught rules or plans • Little situational discrimination (e.g. between safe and unsafe behaviours) • No comparative judgment	• Works directly with NORM-bearing or contaminated objects	• Must be taught basic radiation safety principles, e.g. ALARA • Must be taught to use PPE	**Requires constant supervision** • Task specific
2. Advanced beginner	• Follows guidelines for work aligned to key task attributes or aspects • Some situational discrimination (e.g. safe or unsafe) • All attributes and aspects are treated separately and given equal importance	• May perform routine radiation surveys or sampling, unsupervised • May be specifically assigned to work in higher NORM exposure situations	• Must be taught radiation safety principles in more detail, e.g. radioactivity and radiation, biological effects, dose and risk, limits, use of survey equipment and personal dosimeters	**Works largely unsupervised** • Task specific • Procedural — tasks are concatenated into a coherent process or flow sheet
3. Competent	• Multi-tasking — can also prioritize • Contextualizes routine actions in terms of longer-term goals • Methodical planning with limited adaptability • Differentiates standardized and routine procedures from exceptions • Can diagnose and remedy routine faults • Follows all safety procedures; anticipates and prevents risks	• May supervise teams working in NORM exposure situations • May participate in drafting radiation work permits • May participate in planning for decontamination, decommissioning, or waste disposal activities, including QA–QC	• Must be taught to supervise a team working in a radiation environment • Must be taught to recognize and characterize an exposure situation • Must be taught how to assess dose and manage time, distance and shielding to keep doses ALARA • Must be taught radiological waste management	**Supervises, within defined framework** • Interpersonal — such as communications (oral and written) and teamwork • Contextual — demonstrates capacity to work within the wider operating or process environment • Reporting — outputs

TABLE 1. COMPETENCY MATRIX (cont.)

Level	Generic	NORM operational	(Radiation) safety hurdle	Management
4. Proficient	• Understands situations holistically • Knows quickly what is most important in a situation; reacts instinctively safely • Perceives deviations from the normal pattern and is adaptive • Practised at decision-making • Uses maxims for guidance, whose meaning varies according to situational need, and can direct others	• Will be placed in charge of health physics duties for a site or company to include: — site characterization and monitoring — personal monitoring — shipping — reporting — regulatory compliance • May construct and direct corporate initiatives involving handling of NORM • May author corporate policies, procedures and best practices	• Must acquire skills to function as site or corporate radiation safety officer • Must be able to design radiation safety and environmental monitoring programmes • Must be able to analyse dosimetry and environmental data • Must be able to report analysis results to corporate officers and regulatory agencies	**Manages/decides** • Performance — optimization • Contingent — such as dealing successfully with the unexpected or unforeseen • Accountability — legally liable for radiation protection decisions made on behalf of the company

TABLE 1. COMPETENCY MATRIX (cont.)

Level	Generic	NORM operational	(Radiation) safety hurdle	Management
5. Expert	• No longer relies on rules, guidelines or maxims • Intuitive grasp of situations based on deep tacit understanding • Defines performance and safety outcome measures; can spot emerging trends • Analytic approaches used only in novel situations or when problems occur • Vision of what is possible	• Is capable of adapting existing or developing new policies and procedures for contingent or unforseen events • Is capable of strategic planning and foresight including ‘what if’ modelling and scenario development • Is capable of developing innovative strategies for radiation protection • Will participate in technical dialogue with standard-setting or regulatory agencies • Will develop the corporate vision on how NORM is used, avoided, or otherwise managed	• Must have access to information in order to evaluate NORM impacts for a company or entire industry • Must have expert knowledge of radiation principles and NORM to author policies, procedures and best practices • Must have access to information in order to contribute to organizations that set standards or draft regulations	**Leads** • Defines, evaluates, redefines processes and competencies • Influences scientific debate and regulatory policy • Influences or establishes corporate vision and mission statement

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REVIEW OF THE SYMPOSIUM AND OVERALL CONCLUSIONS

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1. INTRODUCTION

As the final rapporteur, I was asked to give an overview of the whole symposium rather than just a straight summary of the final sessions. In particular, I was asked to try and take a step back from the individual presentations and consider:

(a) The overall themes and conclusions from NORM VI;
(b) The progress made since NORM V, held in Seville in 2007;
(c) Any issues that might be relevant for NORM VII (Kochi, India, 2013).

First, I would like to thank the organizing committees, the symposium sponsors, and the local hosts, all of whom helped to ensure a stimulating and successful event.

The symposium contained oral presentations (invited and contributed) and a wide selection of poster presentations. Certain themes emerged, and I have attempted to summarize these below.

2. NORM STANDARDS AND REGULATION

The need to establish standards, especially standards defining the scope of regulatory control, is a recurring theme at all of the NORM symposia. Furthermore, given that most NORM industries are international, there are clear benefits to be derived from the harmonization of safety standards. At NORM V, there were signs of a common approach developing, based on numerical criteria for entry into, and exit from, regulation as a practice (for example, activity concentrations[1] being above 1 Bq/g) and, in the case of practices, numerical criteria for exemption (for example, annual doses received by workers being below 1 mSv). At this symposium, it was clear that these criteria are now widely accepted as the way forward for future safety standards. For example, the use of

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[1] For radionuclides in the ^{238}U and ^{232}Th decay chains.

1 Bq/g as a level below which regulatory control is unnecessary for most types of material is now referred to in draft revisions of IAEA Safety Series No. 115 and of the European Council Directive 96/29/Euratom. Similarly, both of these draft revisions include provision for exemption from regulatory control when worker doses are below 1 mSv and for the application of a graded approach to regulation above this value.

All of this represents positive progress since NORM V. One cause for concern (as in Seville) is that 1 Bq/g was occasionally referred to as a ‘limit’. This was never the original intention, and such an interpretation can raise significant problems. In fact, under many circumstances 1 Bq/g is a conservative value, and less restrictive activity concentration criteria may beconsidered appropriate.

There are some differences between the draft revisions of the IAEA and European safety standards, for example in the approach to NORM in building materials and the requirements for NORM effluents and residues. It is rather early to assess the full implications of these differences. Nevertheless, the impression from a number of presentations and posters relating to building materials and effluents was that either set of requirements could be accommodated in practice.

The most apparent differences in approach, however, arise from interpreting the ICRP recommendations on exposure situations, in particular defining what are planned exposure situations and what are existing exposure situations. These new recommendations appeared between NORM V and VI, and are clearly open to very diverse interpretations. To add to the confusion, the practical implications of these distinctions are not obvious. A clear message from the symposium was that further consideration of, and guidance on, the application of the ICRP recommendations to NORM was required. To this end, it was reported in the final discussion session that an ICRP Task Group on NORM is planning to address this.

While there was much agreement on exempting practices involving NORM according to the 1 mSv/a (worker dose) criterion, there was little practical information on how to apply a graded regulatory approach in cases where doses are higher. Consequently, there is a danger of regulation being perceived in an excessively negative light. Hopefully, by the time of NORM VII, there will be more examples of how the system can be applied in a proportionate manner.

3. AN INDUSTRY DRIVEN APPROACH

Although standards and regulations are being developed for NORM in general, it is clear that individual NORM industries are very different, as are the practical radiation protection challenges they face. Consequently, solutions that are appropriate for the oil and gas industry may be very different from those appropriate for the phosphate industry, even though both may be dealing with ^{226}Ra and/or ^{228}Ra. To this end, there is increasing evidence of the value of an industry based approach, either through the so-called ‘positive list’ of industries proposed for Europe, or through industry specific publications such as IAEA Safety Reports.

This approach also offers a more direct opportunity for industries to be involved in the development of practical radiation protection regimes. The NORM symposia have always enjoyed and encouraged industry involvement, and this was the case at NORM VI. Despite this, there remains a tangible industry frustration with the slow progress towards a coherent and understandable approach to protection. One lesson from the oil and gas industry is to not wait, but to take the lead in developing and implementing appropriate protection strategies and solutions.

4. OPERATIONAL RADIATION PROTECTION

As with NORM V, there was much information on methods for determining radiation doses for both workers and the public. In considering public exposures such as those from the migration of NORM radionuclides through the environment, the use of exposure models is necessary. There is still a tendency for such models to be very conservative, or even ‘worst case’, which results in improbably high estimated doses. These results are of limited value when considering the optimization of protection, in which the resources expended should be proportional to the benefit gained. Thus, the use of realistic models, with appropriate sensitivity analyses, is to be encouraged.

In terms of worker exposures, the emphasis should be on actual monitoring data (workplace and individual) to ensure that dose estimates are realistic. This would now seem to be the most common approach, although some evidence of pessimistic modelling still remains.

At the symposium, the most common reason for determining individual doses was to establish the need (or otherwise) for regulatory control. In contrast, there was only limited information presented on how such doses might then be optimized in practice. Looking collectively at the information presented, annual doses from work with NORM (excluding radon) may be summarized as:

- <1 mSv in most cases,
- 1–6 mSv in some cases,
- >6 mSv in a very few cases.

This distribution is broadly the same as that seen for work with artificial sources of ionizing radiation. Of course, it could be argued that artificial sources have traditionally been subject to a more stringent regulatory regime. Nevertheless, the data are consistent enough to suggest that a similar protection philosophy — through engineering controls, working procedures, and personal protective equipment — is appropriate. At future NORM events it would be useful to share more information on how this can be applied in different NORM workplaces.

5. INTERNATIONAL ISSUES AND TRANSPORT OF NORM

In the NORM V summary, it was suggested that further efforts to encourage the involvement of key NORM producing and processing countries, such as China, were needed. It was, therefore, pleasing to see such involvement at NORM VI. Furthermore, the important role of the IAEA Technical Cooperation Programme in funding the participation of experts from several countries is very much appreciated.

Regarding transport, a factor of 10 is applied to the NORM activity concentration exemption levels. The appropriateness of this factor has been reviewed in a series of studies of worker and public doses. Although in some cases this figure might be considered too restrictive, the overall results fail to provide a persuasive argument for adjusting the current exemption levels for transport of NORM.

Problems associated with the international transport of radioactive material constituted the main reason for reviewing the exemption levels for transport of NORM. However, from the evidence provided at NORM VI, these problems are not restricted to just those radioactive materials that are subject to the IAEA Transport Regulations. The increasing use of radiation detectors at borders means that NORM, whether exempt or not, is being quarantined, leading to delays or even denial of shipment. Monitoring at borders plays a key role in the safety and security of radioactive sources, and can only be expected to become more common. Increasingly sophisticated monitoring systems allow for the identification and categorization of NORM. This, together with improved operator training, may offer the best means of resolving the current problems.

6. THE MANAGEMENT OF NORM RESIDUES

As with previous symposia, the management of NORM residues remains the main challenge for a number of industries, with the problems very much depending on the properties of the residues themselves. This includes not just current operations, but a significant number of legacy sites, many of which previously operated in accordance with less stringent health, safety and environmental standards.

For some NORM industries, especially those producing low or medium volume residues with higher activity concentrations, a suitable waste disposal repository is the only practicable option. It is increasingly accepted that many NORM residues may be disposed of in a manner similar to that for other hazardous wastes, rather than being consigned to facilities originally designed for high activity radioactive waste (containing radionuclides of artificial origin). The problem is still the lack of suitable facilities prepared to accept NORM residues, although a small number are now beginning to appear.

For the phosphate industry, the main issue is phosphogypsum, which contains relatively low NORM activity concentrations, but is produced in vast quantities. As indicated by several presentations, there are several options for the use of this material that do have to be explored. Many of these options appear promising from a scientific perspective, although there is still work to be done before it can be said that there is universal support for such strategies. Certainly, it is important to continue working on a sound scientific underpinning for the management strategy, and this needs to include a consideration of both radiological and non-radiological hazards. In addition, a process of wider stakeholder engagement is needed in the decision making process. In this respect, there were some interesting discussions on the prevailing public and political mood towards recycling and use of NORM residues in general.

Finally, the ‘dilute and disperse’ option continues to provoke an interesting debate. Despite this option being controversial within the overall context of waste management, it may still be the optimum solution for some NORM residues, especially where it effectively returns radionuclides to their natural state.

7. RADON

This topic received relatively little attention, although a number of posters provided further evidence that exposures from radon can be very substantial. The situation is made more complicated by separately considering radon from NORM and from the existing environment, even though both will ultimately draw upon on the same protection options.

The new ICRP exposure situations are also very significant in respect of radon. Dose reference levels are fundamentally different to the current radon action levels, and it is as yet unclear how to implement optimization in practice below these levels.

8. CLOSING THOUGHTS

At NORM V, a key question was whether there was chaos or consensus in relation to managing NORM — and the answer was that there was still a little of both! There has been some key progress since then, most noticeably in the drafting of revised versions of IAEA Safety Series No. 115 and the European Council Directive 96/29/Euratom, both of which aim to provide a sensible framework for managing NORM. To some extent, the new ICRP recommendations on planned and existing exposure situations have introduced confusion into this process, and further clarification is needed, and is indeed planned.

At the end of NORM , it was suggested that the time had come to move from discussion to implementation. However, it would appear that the process of redrafting standards has mostly filled the gap between Seville and Marrakesh. In the meantime, the industries themselves have continued to search for their own solutions. By the time of NORM VII in 2013, it is hoped that we will have a clear and coherent radiation protection framework, upon which proportionate and effective protection options can be implemented.

Finally, the symposium included many more issues than those discussed above, and it is impossible in this short summary to do justice to all the material presented. However, it is hoped that the above at least gives some flavour of the proceedings. I would again like to say how valuable NORM VI was, and pass my compliments to the organizers, presenters and all those who participated in the discussions.

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All minerals and raw materials contain radionuclides of natural origin. For most human activities involving minerals and raw materials, the levels of exposure to these radionuclides are not significantly greater than normal background levels. However, certain work activities can give rise to significantly enhanced exposures that may need to be controlled by regulation. Material giving rise to these enhanced exposures has become known as naturally occurring radioactive material (NORM). Historically, most regulatory attention has been focused on the mining and processing of uranium ore, because such activities are a direct consequence of the radioactivity in the ore and form part of the nuclear fuel cycle. Over the past decade or two, however, more and more countries have introduced measures to regulate exposures arising from a wider range of natural sources. The NORM VI symposium, which was held in Marrakesh, Morocco, from 22–26 March 2010, provided an important opportunity to review the developments that had taken place during the three year period since the NORM V symposium in 2007. This period, which began with the publication of new radiation protection recommendations by the International Commission on Radiological Protection, was characterized by ongoing activities to revise international standards on radiation protection and safety. These Proceedings contain all 38 papers accepted for oral presentation and four rapporteur reports, as well as a summary that concludes with the main findings of the symposium. Text versions of 43 poster presentations are provided on a CD-ROM which accompanies these Proceedings.

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