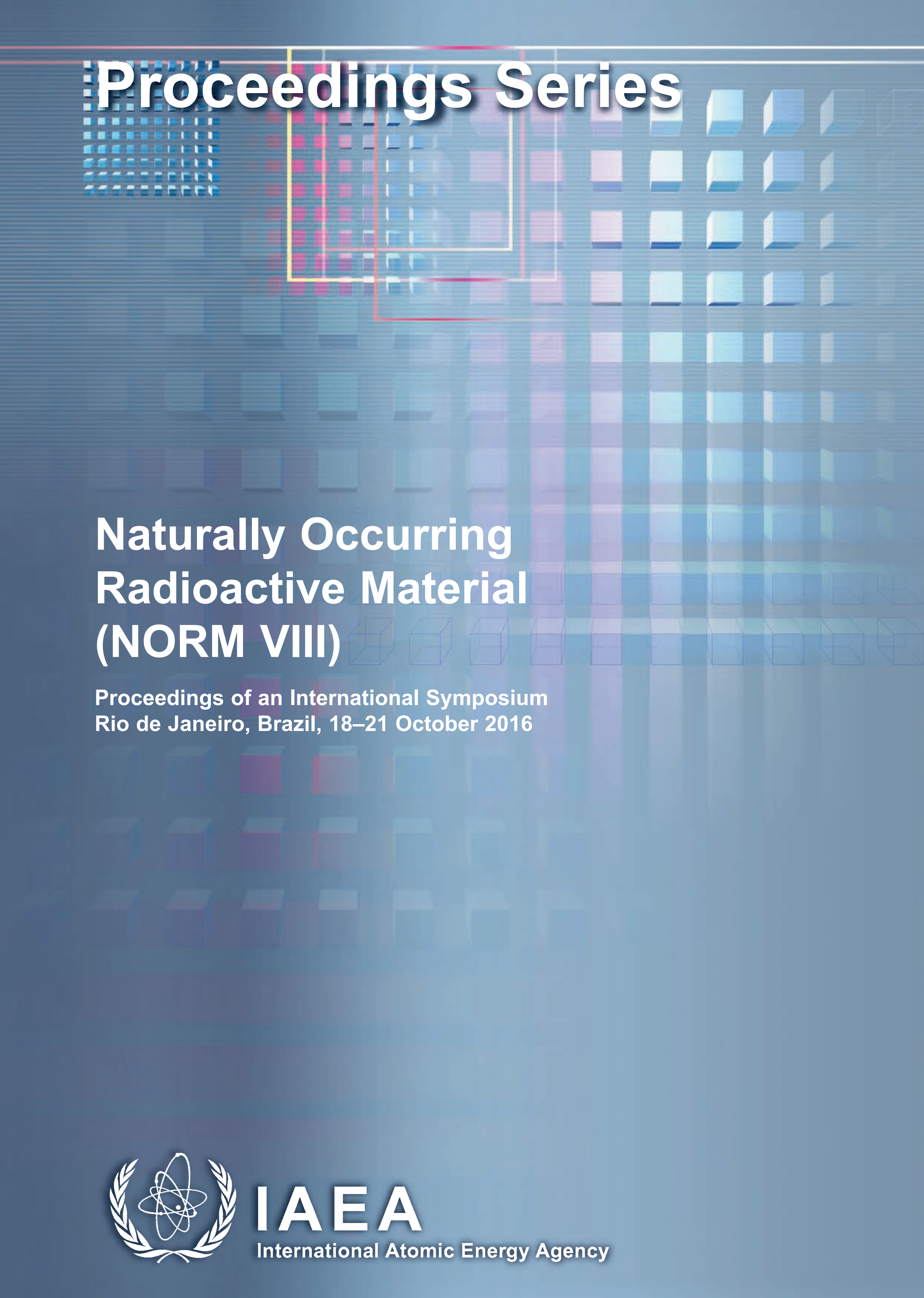

Proceedings Series

Naturally Occurring Radioactive Material (NORM VIII)

Proceedings of an International Symposium
Rio de Janeiro, Brazil, 18–21 October 2016

IAEA
International Atomic Energy Agency

NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM VIII)

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PROCEEDINGS SERIES

NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM VIII)

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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. 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 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).

As a direct consequence of the 1996 European Council Directive 96/29/Euratom 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, the fifth (NORM V) in Seville, Spain, in 2007, the sixth (NORM VI) in Marrakesh, Morocco, in 2010 and the seventh (NORM VII) in Beijing in 2013. In addition, a symposium on Technologically Enhanced Natural Radiation was held in Rio de Janeiro, Brazil, in 1999, reflecting the growing interest at that time 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 symposium in 1999 and the Szczyrk symposium in 2004 were published as IAEA-TECDOC-1271 and IAEA-TECDOC-1472, respectively, while the proceedings of the Seville, Marrakesh and Beijing symposia were published in the IAEA Proceedings Series. In the case of NORM VIII, the IAEA entered into a formal cooperation arrangement with the organizer (as it had done for other recent symposia in the series), in terms of which the IAEA, in addition to publishing these Proceedings, served on the Steering Committee and Scientific Committee and provided financial support to several participants from Member States eligible to receive assistance under the IAEA's technical cooperation programme.

The NORM VIII symposium was attended by 257 participants from 50 countries and provided an important opportunity to review the developments that had taken place during the three year period since the Beijing symposium in 2013. This period was characterized by ongoing activities to implement international standards on radiation protection and safety in many countries. These Proceedings contain 31 papers accepted for oral presentation, text versions of 35 poster presentations and a summary that concludes with the main findings of the symposium.

The IAEA, on behalf of the Brazilian organizers — the Institute of Radiation Protection and Dosimetry, and the National Nuclear Energy Commission — gratefully acknowledges the cooperation and support of all the organizations and individuals who contributed to the success of this symposium.

The IAEA officer responsible for this publication was H.B. Okyar of the Division of Radiation, Transport and Waste Safety.

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SUMMARY

1. BACKGROUND TO THE SYMPOSIUM

1.1. Objectives

This Symposium, the eighth 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, with the overall objectives of addressing radiation protection issues, discussing the results of new research, exploring practical case studies of industrial applications and identifying new societal needs and technical requirements for regulatory bodies and NORM industries. The Symposium provided a platform for experts from NORM industries, academic and research institutions and regulatory bodies from all over the world to share experiences, to identify opportunities, to analyse current challenges, and to review progress made in identifying, quantifying and managing the radiological risks associated with industrial processes involving NORM. Ongoing activities to implement new international standards during the period since the last NORM symposium in 2013 provided an important backdrop to the presentations and discussion.

As with previous symposia in this series, the technical programme was well subscribed. These Proceedings contain 31 papers that were accepted for oral presentation along with 35 contributions in the form of posters.

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 that the implementation of a new European Council Directive[1] 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 European Union Member States and as knowledge about exposure levels 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 mineral processing industry, with large quantities of minerals being mined and beneficiated in countries far 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.

Given this background, it was fitting that Brazil — a major source, processor, user and supplier of industrial minerals, many of which contained elevated concentrations of radionuclides of natural origin — was chosen as the venue for the NORM VIII symposium. The planning of the symposium followed an approach similar to that adopted for NORM V, NORM VI and NORM VII, in that steps were taken to encourage participation from all regions of the world. The steering committee arranged for broad international representation on the scientific committee of the symposium and encouraged members of that committee to actively promote participation in the symposium from within their own geographic regions. Furthermore, the IAEA once again provided financial support to several participants from Member States eligible to receive assistance under the IAEA technical cooperation programme.

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[1] COUNCIL OF THE EUROPEAN UNION, Council 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, Official Journal of the European Communities, L 159, vol. 39 (1996).

These efforts were evidently successful in that the symposium attracted 257 participants from 50 countries. While the high level of participation by individuals from regulatory bodies and scientific institutes was encouraging, it was noted that, once again, there were relatively few participants from NORM industries.

2. RADIATION PROTECTION STANDARDS AND REGULATORY APPROACHES

2.1. Activities of the IAEA and the International Commission on Radiological Protection

The latest version of the IAEA's International Basic Safety Standards — IAEA Safety Standards Series No. GSR Part 3, published in 2014 (the BSS)[2] — establishes radiation protection requirements for NORM industries in planned and existing exposure situations. The IAEA has initiated many activities, including the development of safety guides and safety reports, for assisting Member States in implementing the relevant requirements of the BSS. It was pointed out in this regard that the characteristics of industrial processes and exposure situations involving NORM are in many cases quite different from those associated with other activities involving radioactivity. Reference was made to the wide diversity of industrial processes and process materials, the generally very low (but sometimes unpredictable) exposure levels, the presence of non-radiological hazards that may well be of greater concern than radioactivity, and the involvement of several different regulatory bodies. This situation gives rise to various challenges for operators and regulatory bodies alike. For instance, guidance on radiation protection and NORM residue management has to be tailored to specific NORM industries and/or industrial processes and has to address the management of extremely large amounts of NORM residues (including the use or recycling of residues where possible) and the remediation of contaminated legacy sites. The work programme includes the development of industry specific guidelines for radiation protection and management of NORM residues, long term management of bulk NORM residues and remediation of legacy sites contaminated with NORM. The programme also addresses the need for enhanced and improved levels of knowledge, understanding and communication. It was emphasized that the the IAEA work programme on NORM focuses heavily on the application of the graded approach to regulation, one of the key concepts in the BSS.

It was reported that the International Commission on Radiological Protection had, in 2013, relaunched a task group to prepare a report on exposure to NORM. It was pointed out that industrial activities involving NORM tend to be well-established activities that continue to be regulated by various authorities concerned with the control of non-radiological hazards. Consequently, when dealing with radiological hazards the need for a graded approach to regulation is not always well appreciated and a radiation protection culture is often lacking. In addition, it seems that there is still some confusion as to whether activities involving NORM should be regarded as planned exposure situations or existing exposure situations.

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[2] EUROPEAN ATOMIC ENERGY COMMUNITY, FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS, INTERNATIONAL ATOMIC ENERGY AGENCY, INTERNATIONAL LABOUR ORGANIZATION, OECD NUCLEAR ENERGY AGENCY, PAN AMERICAN HEALTH ORGANIZATION, UNITED NATIONS ENVIRONMENT PROGRAMME, WORLD HEALTH ORGANIZATION, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3, IAEA, Vienna (2014).

2.2. Implementation of international standards

The new International Basic Safety Standards published by the IAEA in 2014 have been implemented within the European Union through a new European Council Directive[3]. With regard to NORM, this Directive has achieved some progress in reducing (but not eliminating) the complications and confusion that existed previously — these had given rise to some misunderstandings and differences in interpretation, resulting in overregulation in some instances. For example, the new Directive has now introduced numerical criteria for NORM with respect to clearance (e.g. 1 Bq/g for activity concentrations of uranium and thorium series radionuclides) and exemption (an annual effective dose of the order of 1 mSv/a). Nevertheless, there is still some confusion with regard to the newly introduced concept of reference levels in existing exposure situations, with the result that such reference levels are sometimes being erroneously treated as action levels or as limits, rather than as criteria for prioritizing the need for protective or remedial actions and below which optimization of protection is still required. Among other observations, it was noted that, for radiological assessment purposes, there is an ongoing need to give more effort to using actual measurements rather than modelling, since exposures to NORM are often difficult to predict reliably by modelling and tend to be overestimated in such cases. It is encouraging to see that by-product use and recycling of NORM residues are steadily gaining acceptance. Where disposal as waste is the only feasible option, more attention needs to be given to the possible use of facilities for general industrial waste (hazardous or non-hazardous) rather than radioactive waste repositories.

The regulation of NORM in the United States of America continues to be complicated, as a result of being primarily (but not entirely) left to individual states. While this approach provides flexibility, it can lead to inconsistencies in regulation and diverse approaches, particularly in the management of NORM residues. Even the terminology can be confusing, since there is no single definition of NORM and widespread use is still made of the additional term TENORM (with such use having largely fallen away in other countries). Inconsistencies between states are now starting to attract more attention, and are even giving rise to legal issues, while there is now a recognized need for more data on exposure to NORM.

In the Netherlands, regulations and procedures for activities involving NORM have been under development for many years, although with the establishment of the new European Directive in 2014 there is some work still to be done in order to implement the new requirements while at the same time reducing the costs involved. Experience over the past years has highlighted a tendency for the regulation of NORM to be unnecessarily restrictive and it is felt that this needs to be relaxed. Experience has also demonstrated the benefits of cooperation between all stakeholders. With regard to NORM residues, the by-product use or recycling of such residues is the primary target of the NORM residue management system. For application in civil engineering, a specific requirement in 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 recycling or use are not feasible can the material be disposed of, and only then is it considered to be waste.

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[3] COUNCIL OF THE EUROPEAN UNION, Council Directive 2013/59/Euratom of 5 Dec 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, Official Journal of the European Communities, L13, vol. 57 (2014).

In Austria, implementation of the latest European Council Directive is now getting underway. It has been found that more data are needed on the various activities involving NORM and the associated exposures. Also, additional guidelines will have to be developed. The implementation process is based firmly on the graded approach to regulation. There are many legacy sites potentially contaminated with radionuclides of natural origin, especially isotopes of radium and thorium. A legacy site catalogue is regularly updated, sites are being characterized and the associated radiological risks are being prioritized on the basis that an activity concentration of 1 Bq/g for uranium and thorium series radionuclides is assumed to be broadly equivalent to an annual dose of 1 mSv. For each site identified as requiring action, a decision will be made on whether to decontaminate or secure the site. Factors influencing this decision include the size of the site, whether it is enclosed or open, the solubility of the contaminant and the possibility of groundwater contamination. The approach for sites that are to be remediated involves a dose assessment, radiation protection and monitoring during the remediation work, coordination of the work carried out by the various experts and companies and a post-remediation survey of environmental media. The local population will be informed of the radiological risks and the waste from the remediation work will be disposed of at a hazardous waste landfill facility or at a radioactive waste repository, depending on the radiological risk, on a case by case basis.

In Brazil, a new regulation based on the latest version of the IAEA BSS has been promulgated. Implementation of this regulation has begun, with the classification of industrial activities involving NORM into one of three risk categories: high, medium and low. The industrial activities concerned are classified according to the activity concentrations of ^{238}U and ^{232}Th and — in the case of very low activity concentrations — according to risk (in terms of dose). It has been found that, of the 19 facilities concerned, nine were classified as low risk and four were classified as medium risk. The six facilities classified as high risk were facilities involved in the production of niobium and tantalite.

It is clear from many of the presentations at the Symposium that worldwide implementation of international standards applicable to NORM and the use of supporting documentation by governments and regulatory bodies continues to grow. However, concern was expressed that NORM industries were not using the guidance and supporting information available at the international level and the participation of NORM industry experts in the NORM symposia had declined to very low levels.

2.3. Transport of NORM

The application of the IAEA Regulations for the Safe Transport of Radioactive Material (the Transport Regulations)[4] to the transport of NORM has given rise to some significant problems. The problems relate principally to international shipments and arise from a lack of understanding and communication between transport companies and government departments in different countries. These problems have resulted in rejections of shipments at ports of entry and claims for compensation by workers and by owners of properties near transport routes. One particular problem area is related to NORM that presents additional, non-radiological hazards such as corrosivity, biological hazards, flammability or chemical toxicity. Other problems arise from the temporary storage of NORM shipments in transit storage areas (regulatory control for material in storage is more restrictive than that for material in transport), the triggering of alarms by shipments containing NORM but which are exempt from the Transport Regulations, the buildup of radon in containers and hulls of ships (which can be significant even for exempt shipments) and a failure to recognize that, for the purposes of defining surface contamination, ^{226}Ra is not a low toxicity alpha emitter and is consequently subject to a surface contamination

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[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Material, 2012 Edition, IAEA Safety Standards Series No.SSR-6, IAEA, Vienna (2012).

limit ten times lower than that for other radionuclides associated with NORM contamination on the surfaces of objects in transport.

Investigation of these problems has highlighted the following contributory factors: a lack of communication between parties concerning legal requirements; misinterpretation of legislation, regulations and guidelines; the use of different editions of the Transport Regulations in various countries; the adoption of national regulations and/or standards that are in conflict with the Transport Regulations; and regulatory inconsistencies between countries concerning the transshipment of NORM through ports on the way to the final destination.

2.4. Use of raw materials and industrial residues for construction

Materials used for construction purposes are mostly obtained from natural raw materials, but may also be associated with residues from the mining and processing of minerals. In both cases, the materials contain radionuclides of natural origin that represent a source of public exposure. For some materials, this exposure may be significantly higher than exposure to normal natural background radiation. In terms of international standards, the use of such materials for construction purposes may be subject to restrictions, depending on the dose to members of the public. This dose generally has to be assessed by exposure modelling based on the activity concentrations of radionuclides of natural origin contained within the material.

In a project funded by the European Union, a large number of worldwide materials used for construction purposes are being investigated. Activity concentrations of individual radionuclides of natural origin have been measured in 30 materials (23 natural materials and 7 industrial residues) and used for the calculation of external gamma exposures of members of the public when used as building materials. For ^{226}Ra, activity concentrations of up to 27.8 Bq/g were reported for natural materials and up to 3.1 Bq/g for industrial residues. For ^{232}Th, the corresponding activity concentrations were up to 0.9 Bq/g for natural materials and up to 1.35 Bq/g for industrial residues. Two approaches to dose assessment were used: the ‘I-index’ screening approach developed within the European Union and a more precise approach developed by the IAEA and published as an IAEA Safety Guide. It was found that the I-index approach tended to overestimate doses, sometimes to a significant extent (up to 70%). This could result in unnecessary regulatory restrictions being placed on the use of many materials for building purposes.

3. ACTIVITY CONCENTRATIONS IN RAW MATERIALS, PROCESS MATERIALS AND PRODUCTS

3.1. Production and use of thorium-containing materials

Heavy-mineral sand deposits tend to be associated with thorium because of the presence of monazite. Two heavy-mineral sand operations in Mozambique producing zircon and rare earth elements were investigated. The activity concentrations in the sand dune top layer, sand waste, magnetic ilmenite and magnetic fraction were all less than 1 Bq/g. The activity concentrations in the remaining materials were consistent with values obtained from other heavy-mineral sand operations around the world:

Sand dune, rich layer:	1.01 Bq/g ^{232}Th	0.4 Bq/g ^{238}U
Wet mineral concentrate:	3.2 and 1.4 Bq/g ^{232}Th	0.7 and 0.6 Bq/g ^{238}U
Non-magnetic fraction:	1.6 and 4.5 Bq/g ^{232}Th	2.7 and 1.6 Bq/g ^{238}U
Zircon:	0.2 Bq/g ^{232}Th	1.1 Bq/g ^{238}U

Activity concentration measurements were performed on thorium-containing gas mantles in Mexico. The ^{232}Th activity concentrations ranged from 232 to 683 Bq/g, consistent with values measured in other countries. While thorium-containing gas mantles have largely been replaced by thorium-free products in various countries, they can still be purchased in Mexico and disposed of in normal household waste.

Commercially available paint contains titanium dioxide as a pigment material. This is produced from heavy-mineral sand which contains thorium in the form of monazite. The pigment production process is essentially a purification process, so the final product is not expected to contain significant amounts of thorium. In Brazil, the activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K were measured in several samples of wall paint. As would be expected, none of the activity concentrations were significantly elevated.

3.2. Oil and gas production

In the processing of crude oil in a refinery, over 1000 t of residues containing radionuclides of natural origin can be generated in a year. The use or disposal of these residues is subject to limits on activity concentration. Measurements performed on various samples collected from an oil refinery showed that all of the sludge samples were below 1 Bq/g but high ^{210}Pb activities were measured on heat exchanger scale (up to 8 Bq/g) and installed equipment in distillation columns (up to 25 Bq/g). In Germany the 95 % upper confidence limit of the mean of a random sample activity concentration has to fall below 1 Bq/g concerning the dumping or combustion of NORM residues. Several samples were taken from sewage sludge (800 t/a) and oil sludge (500 t/a) as independent random samples. From these samples the upper confidence limit for the confidence level 0.95 was determined by means of both classical and modern numerical (‘bootstrapping’) statistical methods. In the case of ten independently gathered samples of oil sludge, the control limit was exceeded in terms of both statistical methods. When the sample size was expanded to 20 samples, it could be shown by means of the bootstrap method that the control limit was not exceeded. By using the classical method, the control limit was exceeded again as previously.

The deposition of ^{210}Pb, particularly in gas production facilities, was also highlighted from the point of view that there are two distinct deposition mechanisms. The decay of ^{222}Rn can generate unsupported ^{210}Pb in association with dispersed particles and colloids. It can also generate supported ^{210}Pb in the form of very thin deposits that are invisible and difficult to detect. In the latter case, activity concentrations exceeding 1 Bq/g are commonly found.

A survey of oil production facilities in Ukraine identified the presence of sludge and other waste with radionuclide activity concentrations far in excess of 1 Bq/g. The mean activity concentrations for a given facility were up to 8 Bq/g for ^{238}U, up to 9 Bq/g for ^{226}Ra, up to 11 Bq/g for ^{210}Pb and up to 3 Bq/g for ^{232}Th. Several oil refineries were also surveyed. Samples from waste disposal ponds were analysed and the average radionuclide activity concentrations were found to be 5 Bq/g for ^{238}U and ^{226}Ra, 7 Bq/g for ^{210}Pb and 2 Bq/g for ^{232}Th.

Activity concentration data for produced water and scale were reported from oil production facilities in Ghana. As expected, the dominant radionuclides were ^{226}Ra and ^{228}Ra. Activity concentrations of these radionuclides in produced water ranged from 6 to 34 Bq/L, while in scale the activity concentrations ranged from 27 to 58 Bq/g. These results were consistent with values reported from elsewhere in the world.

Activity concentrations of radionuclides in the uranium decay series and thorium decay series were measured in soil samples gathered from around oil extraction sites in Kuwait and Qatar. The concentrations were all significantly below 1 Bq/g. While the results showed some evidence of elevated concentrations at these locations, the upper bound of the normal worldwide range for rocks and soil was exceeded only in the case of one sample.

3.3. Phosphate production

The following activity concentration data were reported for phosphate production facilities in various parts of the world:

(a) A phosphoric acid plant in Zimbabwe had suffered a decline in production in recent years and process materials and residues from past production were stored at the site. Contrary to expectations, the activity concentrations in the various process materials were not significantly enhanced except for some radium scale which had an activity concentration of 3 Bq/g.
(b) A phosphate mine and processing facility in Senegal was investigated. The ^{226}Ra concentrations in the phosphate rock, wet phosphate raw material and phosphogypsum were 1.2, 1.1 and 0.6 Bq/g (dry weight), respectively.
(c) A survey of a phosphate fertilizer production facility in Ukraine revealed that the radionuclide activity concentrations in the various process materials were all less than 1 Bq/g.
(d) In Finland, the mining of a phosphate deposit with uranium and thorium mineralization is planned. The average activity concentrations in the deposit are 0.3 Bq/g for ^{238}U and 0.5 Bq/g for ^{232}Th.

3.4. Non-uranium mines

At a tantalite mine and processing facility in Ethiopia, the activity concentration of ^{238}U was found to be about 0.1 Bq/g in unprocessed materials and 0.1–0.3 Bq/g in the tailings. In contrast, the tantalite concentrate had ^{238}U and ^{232}Th activity concentrations of 53 and 36 Bq/g, respectively.

Most rare earth elements are obtained from monazite, a mineral normally associated with a high thorium content. In Spain, a rare earths deposit comprises a particular form of monazite that does not contain high concentrations of thorium. The mining of this deposit was the subject of a radiological investigation. The mined materials were analysed for uranium and thorium series radionuclides, as well as ^{40}K. All activity concentrations were found to be less than 1 Bq/g.

In Brazil, analyses were conducted on samples of rock, soil and underground water from eight underground mines producing agalmatolite, coal, emeralds, fluorite, scheelite, tourmaline and zinc. The mean activity concentration in the rock and soil samples from each mine ranged from 0.005 to 0.34 Bq/g for ^{226}Ra, from 0.005 to 0.13 Bq/g for ^{232}Th and from 0.19 to 1.3 Bq/g for ^{40}K. The mean ^{222}Rn activity concentration in underground water ranged from 2 to 487 Bq/L.

In Finland, mineral deposits containing various metals of value are being mined or are being considered for mining. All the deposits have some uranium mineralization:

(1)	Nickel–zinc–copper–cobalt deposit:	0.0017 wt% U (0.2 B/g ^{238}U),
	Associated gypsum pond waste:	0.058–3.375 Bq/g ^{238}U;
(2)	Gold–cobalt (two deposits):	158 ppm U (2 Bq/g ^{238}U) and 194–347 ppm U (2.3–4.3 Bq/g ^{238}U);
(3)	Gold deposit:	Up to tens of wt% U (≈4000 Bq/g ^{238}U);
(4)	Niobium–rare earths deposit:	Hundreds of ppm U and Th (≈15 Bq/g ^{238}U).

In Ukraine, samples of material were gathered at two iron ore production sites that between them incorporate six mines. Activity concentrations of uranium and thorium series radionuclides were determined. The maximum activity concentration was for ^{210}Pb, with a value of 0.11 Bq/g. The maximum concentrations of other radionuclides were less than half this value. Activity concentrations were also measured on waste samples from non-ferrous metal mining. Again, the maximum activity concentration was for ^{210}Pb, with a value of 0.044 Bq/g, while values for other radionuclides were very much lower.

3.7. Coal fired electricity generation

Ash from the coal burning process was sampled at three points within a generating plant in Turkey. The activity concentrations ranged from 0.15 to 1.15 Bq/g for uranium series radionuclides and from 0.065 to 0.16 Bq/g for thorium series radionuclides. As expected, the highest concentrations of ^{210}Pb were found in flyash in the electrostatic precipitators. Using the ‘I-index’ screening approach widely adopted within the European Union, it was determined that the flyash could be used at proportions of up to 40% in cement without the risk of annual effective doses exceeding 1 mSv.

4. OCCUPATIONAL RADIATION PROTECTION

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has gathered data on the annual effective doses received by workers in activities involving NORM. These data are currently being updated. In the meantime, the latest available information from UNSCEAR, published in 2008, is based on data gathered up to 2002, mostly for workers in mines. This information indicates that the average annual effective dose is about 3 mSv.

4.1. Uranium production facilities

The production of uranium is a major NORM industrial activity worldwide which, after a decline in recent years, is now growing once more. An IAEA Safety Report has been developed, dealing with occupational radiation protection in uranium production facilities. This report is aimed at assisting regulatory bodies and facility operators in implementing a graded approach to the regulation of such activities and generating a common understanding between regulatory bodies, operators, workers and their representatives, and health and safety professionals. The report provides information on the various uranium production processes, the associated radiological hazards, dose assessment methods and the establishment of the overall radiation protection programme.

The IAEA has also established the Uranium Mining Exposure (UMEX) project, the general aim of which is to strengthen and enhance the radiation protection of uranium production workers. More specific aims are to increase the opportunities for optimization of protection and to support quality assurance programmes across the industry. The key activities are: the development of an information system for occupational exposure; the evaluation of the current occupational radiation protection situation; and the identification of instances of good practice, opportunities for improvement and, where appropriate, actions to be implemented for assisting employers, workers, regulatory bodies and other stakeholders in implementing the principle of optimization of protection and safety. In 2012, a questionnaire was distributed to uranium producing countries worldwide. Responses were received from 36 operating companies in 20 countries, together representing about 85% of world uranium production. The responses provided information on current practices for monitoring and reporting of occupational exposure and on occupational exposures determined for 2012. The UMEX project has provided a significant amount of information on the monitoring of worker exposures in uranium production facilities worldwide and on the doses received by workers. The information on monitoring approaches can be summarized as follows:

(a) The determination of external gamma exposure is carried out mostly by individual monitoring of all exposed workers using thermo-luminescent detectors (TLDs). However, only about half of the facilities subtract background radiation, resulting in an overestimation of the exposure in such cases.
(b) Internal exposure to inhaled long-lived radionuclides in airborne dust is determined mostly by area monitoring, although significant use of personal monitoring is made. Time variations in exposure are taken into account mostly through the use of periodic monitoring. The inhaled activity is measured mostly by gross alpha counting of the captured dust particles. Where the exposure time needs to be determined as part of the dose assessment process, this is done mostly through the use of time sheets. Bioassay techniques are not widely used for routine dose assessment. The dose conversion factors used for dose assessment vary significantly between facilities.
(c) Internal exposure to inhaled short-lived radon progeny is determined mostly by monitoring of workgroups using radon progeny monitoring devices. The exposure time is established using time sheets and individual doses are assessed by assigning the workgroup average values.

The information obtained from the UMEX project shows that the average annual effective dose received by workers across uranium production facilities worldwide, weighted according to the number of workers at each facility, is less than 5 mSv. This is consistent with the data provided by UNSCEAR, as described above. It is considered unlikely that a worker will receive an annual dose of more than 15 mSv. For workers in underground uranium mines, most of the dose is received from external exposure to gamma radiation and internal exposure from the inhalation of short-lived radon progeny, with similar contributions from each. For workers in opencast uranium mines and in situ uranium recovery operations, most of the dose originates from external gamma radiation. For workers in uranium ore processing facilities, the majority of the dose originates from internal exposure due to the inhalation of long-lived radionuclides in airborne dust. The results for the various facilities highlight the need for consistency in deciding which workers to monitor. The monitoring of all workers at the facility, regardless of whether they received significant exposure, significantly affects the characteristics of the dose distribution, since the vast majority of workers receive exposures of no real concern and the average dose is not necessarily representative of the true situation. It was demonstrated that the compilation of a ‘normalized’ dose distribution (by dividing the number of workers in each dose range by the total number of workers) gives a more meaningful result that conforms closely to the expected lognormal distribution. The importance of selecting the most appropriate dose interval for the distribution was also highlighted.

4.2. Other industrial facilities

An assessment of occupational exposures in eight non-uranium underground mines in Brazil revealed that internal exposure from the inhalation of short-lived radon progeny was the predominant contributor to the total annual effective dose. Under operating conditions (when the ventilation system was working) the annual effective dose averaged over each of seven mines, estimated from measurements of radon and its progeny over 48 h periods, ranged from 0.2 to 7 mSv. Measurements were also conducted in two mines in which the ventilation system was not working. These gave in each case an annual average effective dose of 21 mSv. A wide range of equilibrium factors between radon and its progeny — from 0.2 to 0.7 — was found, illustrating the shortcomings of using a single default value for dose assessment purposes. Annual effective doses due to external exposure to gamma radiation, determined from dose rate measurements, ranged from 0.1 to 2 mSv.

A preliminary investigation of an opencast tantalite mine in Ethiopia indicated that workers could receive annual effective doses exceeding 1 mSv in the packaging, loading and transport areas, especially near piles of tantalite concentrate. This result reflected the relatively high activity concentrations of uranium and thorium series radionuclides measured in the tantalite concentrate and the very low concentrations in the other process materials.

At a phosphate mine and processing facility in Senegal, the annual effective doses received by workers (assessed assuming an annual exposure time of 2000 h) were found to be 2–4 mSv at most work stations but 16 mSv at the filtration unit and up to 24 mSv close to the surfaces of scaled pipes.

At a phosphoric acid plant in Zimbabwe, gamma dose rates were found not to exceed 0.22 µSv/h, indicating that the annual effective doses received by workers from external exposure would be less than 1 mSv.

During a radiological investigation of a landfill disposal site for NORM waste from titanium dioxide production, the modelling of worker exposures to external gamma radiation predicted that each worker would receive an annual dose of 1.9 mSv, of which 85% was due to external gamma exposure. Subsequently, the exposures were redetermined by on-site observations and measurements. This resulted in a very different outcome. It was found that there was no significant possibility of internal exposure, so 100% of the exposure could be attributed to external gamma radiation. This external exposure was then determined from a gamma survey of the site and the maximum annual worker dose was calculated to be 0.8 mSv. Next, the workers were monitored individually using electronic dosimeters, from which the maximum annual dose was determined to be 0.6 mSv. Finally, the workers were monitored over a 3 month period using TLDs and this gave a maximum annual dose of 0.4 mSv — about 20% of the dose initially determined using predictive modelling. This clearly demonstrates the shortcomings of relying on modelling techniques for assessing doses received by workers.

5. DECOMMISSIONING AND REMEDIATION

5.1. Uranium production

Several presentations provided information on the decommissioning and remediation of uranium production facilities, which can be summarized as follows:

(a) An update was given of the remediation activities by the company Wismut with respect to former uranium production facilities in Germany. After 25 years of work, another 10 years were needed for remaining physical work, beyond which several more years of water treatment and monitoring would be required. Although IAEA standards were generally being followed, more restrictive measures were sometimes being applied. Large reductions in the environmental impact had been achieved. The residues being generated included water treatment residues and contaminated scrap metal, rubble and soil. These residues were being used or recycled where possible but had to be disposed of as waste where necessary. The recycling of scrap metal was actually providing some economic return.

(b) Brazil's first uranium production facility, located on the Poços de Caldas plateau, was in production from 1982 to 1995 and is now undergoing active maintenance prior to being decommissioned. The presence of sulphides in the open pit, waste rock piles and tailings deposits is generating acid mine drainage. This has resulted in significant elevations of uranium and other metals in and around the facility. A uranium concentration of 53 Bq/g was measured in sediment at a water discharge point. Elevated concentrations were also found at points downstream, although significant attenuation was observed. A hydrogeochemical and isotopic study of water sampled from 20 sampling points, together with simulation studies, has provided a large amount of information on the situation and the processes involved, including sulphate reducing bacterial action and the mobility of uranium. The groundwater has been identified as being calcic and sulphated. The main elements and their ionic form, as well as the major dissolved and precipitated minerals have been identified. The mechanisms of pyrite oxidation have been evaluated. A review of current measures for effluent control may be necessary. Preliminary results of laboratory studies were also reported. The use of limestone as a permeable barrier component demonstrated its effectiveness in reducing uranium and sulphate mobility, while the use of alkaline leaching of effluent samples achieved an 88% recovery of uranium from the effluent.

5.2. Rare earths extraction from monazite

In Malaysia, following the decommissioning of a plant for extracting rare earth elements from monazite, the site was remediated in two stages. First, the plant site was rehabilitated and then the NORM waste from the decommissioning and rehabilitation operations was disposed of in a specially constructed near-surface repository consisting of two engineered cells. The first cell was used for the disposal of decomissioning waste (contaminated soil and construction material) while the second cell was used for disposal of thorium hydroxide waste in the form of sludge after it had been stored in a temporary facility nearby. It was packaged in concrete containers. After decomissioning the temporary storage facility, the site was rehabilitated using residual activity concentration criteria of 1 Bq/g for uranium and thorium series radionuclides and 10 Bq/g for ^{40}K, using dilution with clean soil to meet these criteria where necessary, so as to qualify for free release. On completion of the work, a single cap was constructed over both cells. Post-release monitoring of the site for gamma radiation and radioactivity in soil, water and air was carried out. The environment around the disposal site is being monitored for 2 years and institutional control will be maintained for at least 300 years.

6. MANAGEMENT OF NORM RESIDUES

6.1. General issues

The costs and liabilities associated with NORM residue management have not always been properly taken into account over the entire life cycle of mining and minerals processing facilities, especially where the radiological and environmental impacts extend far into the future. Modern financial accounting and reporting tools such as the System of Environmental–Economic Accounting and Full Cost Accounting are now able to capture the true costs of mining and minerals processing activities. It has been demonstrated that, when the full life cycle costs are taken into account, the industry has clearly failed to maintain its former economic performance, as illustrated by a decline of 60% on the stock market in the past 15 years. The underlying reason for this failure seems to be the business model itself. A new approach is needed which encompasses, among other things, an environmentally sustainable, socially acceptable, affordable solution to the problem of residues.

The amount of residues generated run into many billons of tonnes. For instance, uranium production generates almost 100% 'waste'. The corresponding percentage for phosphate production is much lower, at 25–30% of the deposit, but during chemical digestion as part of the so-called wet process the production of 1 t of phosphoric acid generates 5 t of phosphogypsum (which is still widely regarded as waste). The volume of residues is not the only problem. Regulatory bodies, as well as the public, have become fearful of the NORM legacy content, with the result that such residues are often regulated as hazardous waste even if that position contradicts the weight of scientific evidence.

To address these problems, mining and minerals processing residues should no longer be automatically referred to as waste — this important statement was repeated many times during the Symposium. Avoidance of the term 'waste' opens the door to value-add uses based on two complementary processing strategies:

(i) 'Comprehensive extraction' considers an orebody (or even a whole geological basin) as a single, complex resource and seeks to optimize returns from all co-located resources, not a single target.
(ii) 'Zero waste' considers the role of innovation, technology and efficient use of all resources in avoiding the generation of waste; an importent part of this strategy is the use of residues as co-products.

Citing the phosphate industry as an example, attention was drawn to the importance of recovering uranium and rare earth elements from phosphoric acid production and of treating phosphogypsum as a valuable co-product rather than as an ongoing liability (i.e. waste). While the recycling or by-product use of a NORM residue might not always be feasible at the time of its generation, it was pointed out that this situation could change in the future and consideration should be given to the possibility of storing the residue in the meantime, rather than deciding there and then to dispose of it as waste.

The dilution of a NORM residue to reduce its activity concentration may open up possibilities for its recycling or by-product use and thus avoid the need for its disposal as waste. This is in line with the established principal of 'dilute and disperse'. In some situations, however, insufficient material is available for dilution and there is not always a suitable market for the co-product. Furthermore, the application of the dilute and disperse principal to NORM residues still faces considerable opposition from regulatory bodies, environmental groups and members of the public. Further effort is needed to encourage the use of the dilution option where appropriate.

Management of NORM residues requires an assessment of short term human exposure as well as a demonstration that environmental criteria for long term human health protection are met. Because of the wide diversity of NORM industrial activities, risk assessments tend to be designed on a site specific basis. This leads to inconsistencies, especially in situations where previous radiological experience is lacking. To address this problem, a unified approach to environmental impact asessment is now proposed. It is argued that the situation is not as complicated as it might initially appear and that the environmental impacts can be grouped into just three basic scenarios: (i) disposal of large amounts of solid residues on relatively small areas of land, (ii) discharge of contaminated water to water bodies, and (iii) discharge of contaminated gases or airborne dust to the atmosphere. In the case of solid residue deposits of on land, the environmental impact is limited and localized and the affected environment is already irrevocably altered. In the case of liquid and gaseous discharges, the impact may be more widespread and serious. This unified approach to risk assessment could ensure greater consistency between facilities and will make it easier to define relevant criteria.

6.2. Disposal of residue from titanium dioxide production

In the United Kingdom, a titanium dioxide production facility generates more than 100 000 t/a of a calcium based reaction residue. Most of the activity content is associated with radionuclides in the ^{232}Th decay series. The sum of the highest radionuclide activity concentrations in each of the ^{232}Th and ^{238}U series is 1–3 Bq/g. There are currently no options for the use or recycling of this residue, and instead it is disposed of as waste at a non-hazardous waste landfill site. The activity concentration is such that the waste falls within the scope of the applicable legislation and would be either subject to licensing or exemption. A dose assessment was needed in order to determine whether the waste qualified for exemption. The dose criteria are 0.3 mSv/a for members of the public, 1 mSv/a for landfill workers and 3 mSv/a for inadvertent intrusion. For the inadvertent intrusion scenario, where houses are assumed to be built on the site 50 years after closure, the highest dose calculated for the public was 1.3 mSv/a (65% of the dose being from inhalation of radon). This dose is lower than the dose criterion for inadvertent intrusion of 3 mSv/a. Doses to the public from the other scenarios were many orders of magnitude lower than this and considerably lower than the dose criterion of 0.3 mSv/a for members of the public. The maximum dose received by a landfill worker was 0.4 mSv/a. While this was comfortably below the 1 mSv/a dose criterion, a site specific dose assessment was necessary to demonstrate this (a generic assessment based on modelling gave a dose of 1.9 mSv/a). Consequently, the waste was exempted from licensing and subject only to certain regulatory requirements such as record keeping.

6.3. Use of NORM residues in construction materials

In a review by UNSCEAR of exposures received by members of the public due to releases of NORM from industrial activities in the United Kingdom, the exposures were found to have arisen mainly from the use of residues such as flyash and slag in building materials Although the data gathered so far are sparse, it appears that the annual effective doses received are generally less than 1 mSv, but there are instances where this value might be exceeded. An update of public exposure data is envisaged.

A study in Portugal on the use of phosphogypsum in cement as a component of concrete has shown that the heating applied during the cement production process causes radium and polonium to volatilize, while uranium and thorium remain immobilized. It was conluded that the radiological risk associated with the use of this cement for making concrete was low.

7. STAKEHOLDER INVOLVEMENT

Many of the presentations at the Symposium made reference to the importance of stakeholder involvement in the development and implementation of legislation and regulations concerning natural sources of radiation as well as in the planning, operation and decommissioning of industrial activities associated with NORM. In the Netherlands, during the process of developing and implementing legislation on natural sources of radiation over a period of more than 40 years, discussions between all stakeholders had been maintained and the stakeholders were reported as still being cooperative and having confidence in each other and in the process. With regard to industrial facilities involving NORM, it was now a reality that such activities not only had to be acceptable from the point of view of health and safety (and more recently, environmental sustainability), they also had to be socially acceptable. The siting of mine residue deposits close to urban areas and the disposal of mine residues by the ‘dilute and disperse’ approach were examples of approaches that were often not permitted by regulatory bodies — even when demonstrated as having insignificant health and environmental impacts — because, in the absence of adequate stakeholder involvement, they faced opposition by members of the public.

There was growing acceptance of the benefits of, and necessity for, stakeholder involvement and that such involvement needed to be strengthened. However, it was pointed out that efforts to communicate with members of the public have not always been successful, resulting in ongoing misconceptions. This was a very challenging task, but there were indications that it could be made to work. In addition to better communication, the importance of early stakeholder involvement and inclusive decision making was emphasized.

A case study reported from Brazil provided a good illustration of the situation, even though it did not relate specifically to NORM. Attempts had been made to communicate with the local population concerning the implementation of a reactor facility for research and isotope production, but information and awareness within the community remained seriously lacking and the future benefits to the area and local community were not appreciated. It was concluded that the communication approach had focused too much on scientific and technical explanations that were not well understood by the general public and therefore had not addressed the widespread misgivings about the project. It was concluded that further work was needed to provide more information that was readily understandable. In particular, attention should be given to establishing constructive dialogue with the media, since they could exert a powerful influence over public opinion.

8. MAIN FINDINGS OF THE SYMPOSIUM

The main findings listed below are based on the presentations made at the Symposium and also on the discussions around those presentations.

8.1. Regulatory aspects

(a) Encouraging progress is being made around the world in implementing the relevant requirements of the latest version of the IAEA International Basic Safety Standards, as they apply to activities involving NORM. Standards, regulatory approaches, guidance and understanding are becoming more consistent between countries, although many differences still exist.

(b) The requirements for NORM are clearer and more explicit than those published previously and are generally well understood, but some aspects are still giving rise to confusion and/or misinterpretation. Further clarification of the system of radiation protection for NORM and its implementation in practice is needed. Guidance is needed on practical applications using case studies as examples.

(c) The IAEA’s work programme on NORM continues to provide guidance and information on the implementation of the new requirements.

(d) The implementation process at the national level is becoming increasingly based on the graded approach to regulation and better use is being made of the regulatory option of exemption. Regulatory systems for NORM are becoming more robust and based to an increasing extent on well structured policies and strategies. These developments should help to avoid overregulation and reduce costs (a source of concern in recent years).

(e) Concern has been expressed that NORM industries are not making use of the guidance and supporting information available at the international level on the implementation of standards for NORM and are no longer participating significantly in international events such as the NORM symposia. A closer working relationship with industry is needed.
(f) There are ongoing problems with the practical implementation of the IAEA Transport Regulations with respect to international shipments. Contributory factors include poor communication between parties concerning legal requirements; misinterpretation of legal requirements; the use of different editions of the Transport Regulations; national regulations in conflict with the Transport Regulations; and regulatory inconsistencies between countries concerning the transshipment of NORM through intermediate ports. It has been suggested that a specific technical report on these issues should be developed by the IAEA.

8.2. Industrial activities involving NORM

(a) More data have become available on the activity concentrations of radionuclides of natural origin in raw materials, process materials and products, adding to the large database of information that already exists.
(b) Similarly, more data have become available on exposures, particularly occupational exposures, in various mining and minerals processing facilities.
(c) The new data are generally in agreement with the data already in existence, although in many instances (sometimes quite unusually) the activity concentrations and the resulting exposures have been found to be at the very low end of the expected range and thus of no regulatory concern.
(d) No new types of industrial activity involving NORM have come to light.
(e) While measurement techniques are becoming well developed, the interpretation and assessment of measured data is suffering from a lack of harmonization between groups of countries with different needs and agendas. When providing technical support in this regard, there should be a clear understanding of a country's needs.
(f) The Symposium highlighted once again the tendency to overestimate doses arising from industrial facilities involving NORM, for instance by relying on conservative exposure modelling rather than measurements on site, by adopting assessment approaches that are too simplistic, or by not taking into account the contribution of background radiation.

8.3. Management of NORM residues

(a) When the mining and minerals processing industry is assessed using modern financial accounting and reporting systems that take into account the full life cycle costs and liabilities associated with environmental sustainability and social acceptability, it is evident that the industry has failed to maintain its former economic performance. A new approach is needed which encompasses, among other things, an environmentally sustainable, socially acceptable, affordable solution to the problem of residues.
(b) These residues should not be regarded as waste — rather, the new concepts of 'comprehensive extraction' and 'zero waste' should be embraced, in terms of which the returns from all co-located resources in the vicinity of the operation are optimized and the generation of waste is avoided through innovation, technology and efficient use of resources.
(c) A key part of this strategy is the recycling or by-product use of residues. It was evident during the Symposium that the use of this approach for NORM residues is slowly but surely gaining acceptance among governments, regulatory bodies, facility operators and other interested parties.

(d) With respect to the dilution of NORM residues with lower activity material to render them safe for recycling or by-product use (or to facilitate their disposal as industrial waste rather than radioactive waste), the general attitude at the moment seems to be that such an approach is not encouraged and is allowed only on a case by case basis, subject to a radiological impact assessment. This attitude is now being questioned, with the suggestion that the use of dilution in appropriate cicumstances should actually be encouraged, even though efforts might be needed to convince some regulatory bodies and environmental groups that this approach was in best the interests of society (through a reduction in the hazards and liabilities associated with the disposal of enormous amounts of NORM waste).

(e) In the case of NORM residues used as construction materials, it was questioned whether such use was justified since, although it might be in the best interests of facility operators, it did not necessarily provide any benefit to members of the public. However, it had been demonstrated that dilution with lower activity materials ensured that doses received by members of the public would be very small, even when the material was used to construct homes. In view of the very large amounts of NORM residues that might be used as construction materials, it was pointed out that much of it would in any case have to be used for structures such as dikes, dams and roads, for which there was little chance of any significant public exposure.

(f) When NORM residues have to be disposed of as waste, greater attention should be given to the possibility of using facilites for disposal of industrial waste (hazardous or non-hazardous) rather than facilities for the disposal of radioactive waste.

(g) It was concluded that, in view of the fundamental nature of many of the issues in the management of NORM residues, some of them requiring major shifts in policies and attitudes, these issues cannot be expected to be completely resolved in just a few years.

8.4. Stakeholder involvement

(a) There was growing acceptance of the benefits of, and necessity for, stakeholder involvement and that such involvement needed to be strengthened.

(b) Efforts to communicate with members of the public have not always been successful, resulting in ongoing misconceptions. It was clear that this was a very challenging task, but there were indications that such efforts could (and should) be made to work.

INTERNATIONAL AND NATIONAL STANDARDS ON NORM — PRACTICAL IMPLICATIONS

(Session 1)

Chairperson

N. TSURIKOV
Australia

Invited Paper

IAEA WORK PROGRAMME ON NORM: ACHIEVEMENTS AND CHALLENGES

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Abstract

The paper highlights some of the key features of the IAEA's work programme that has been initiated to assist Member States in the implementation of the latest safety requirements for NORM industries, as set out in the IAEA International Basic Safety Standards. The characteristics of industrial processes and exposure situations involving NORM are in many cases quite different from those associated with other activities involving radioactivity. Examples given in the paper refer to the wide diversity of industrial processes and process materials, the generally very low (but sometimes unpredictable) exposure levels, the presence of non-radiological hazards that may well be of greater concern than radioactivity, and the involvement of several different regulatory bodies. This situation gives rise to various challenges for operators and regulatory bodies alike. For instance, guidance on radiation protection and NORM residue management has to be tailored to specific NORM industries and/or industrial processes and has to address the management of extremely large amounts of NORM residues (including the use or recycling of residues where possible) and the remediation of contaminated legacy sites. Reference is also made to the general lack of radiological knowledge and radiation safety culture, as well as the need for engagement with all the various stakeholders.

1. WORK PROGRAMME

The latest version of the IAEA's International Basic Safety Standards — IAEA Safety Standards Series No. GSR Part 3, published in 2014 (the "BSS")[5] — establishes requirements for NORM industries in planned and existing exposure situations. These requirements create new challenges for regulatory bodies and operators because the activities concerned cover broad industrial sectors with highly diversified characteristics and because, in the past, many such activities have not been regulated in terms of radiation safety. In the application of the BSS to NORM activities, consideration needs to be given to radiation protection of workers, the public and the environment for a wide range of NORM industries on a global basis. The IAEA has initiated many activities, including the development of safety guides and safety reports, for assisting Member States in implementing the relevant requirements of the BSS.

In managing NORM residues (including those to be disposed of as NORM waste), radiation protection of the public and environment need to be addressed. This includes the control of NORM discharges, the use and recycling of residues and the application of the concepts of exemption and clearance. The long term safety of NORM waste is a particular challenge, requiring appropriate control of the disposal of such waste in, for instance, conventional landfill facilities and the decommissioning and remediation of relevant facilities and sites. Various options should be considered, understood, applied and assessed as to whether

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[5] EUROPEAN ATOMIC ENERGY COMMUNITY, FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS, INTERNATIONAL ATOMIC ENERGY AGENCY, INTERNATIONAL LABOUR ORGANIZATION, OECD NUCLEAR ENERGY AGENCY, PAN AMERICAN HEALTH ORGANIZATION, UNITED NATIONS ENVIRONMENT PROGRAMME, WORLD HEALTH ORGANIZATION, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3, IAEA, Vienna (2014).

radionuclides and other potentially hazardous materials are appropriately isolated from the human environment.

While the radiological aspects of NORM industries can be characterized in a general way, radiation protection needs to be managed in a site specific and practice specific manner. Thus, radiation protection addressing public health concerns associated with releases of radionuclides to the environment, the incorporation of process residues into building materials, and the safe long term management of NORM residues (including NORM waste) requires tailored and graded approaches and an understanding of consequence management. This in turn requires consultation and engagement with the various stakeholders.

The IAEA work programme on NORM focuses heavily on the application of the graded approach, one of the key concepts in the BSS. The programme includes the development of industry specific guidelines for radiation protection and management of NORM residues, long term management of bulk NORM residues and remediation of legacy sites contaminated with NORM. The work programme also addresses the need for enhanced and improved levels of knowledge, understanding and communication.

2. OPPORTUNITIES AND CHALLENGES

NORM is a crosscutting activity within the IAEA and the intention is to extend the IAEA's library of NORM standards with direct and clear messages to Member States. One of the pillars of such a work programme is worker protection in different NORM industries, which generally needs to deal with multi-hazard situations where the radiological risk is generally not the dominant risk. Referring to the various safety reports published by the IAEA in this technical area, the overarching issues common to all industry sectors are the following:

- Doses received by workers are always expected to be below thresholds for deterministic effects — indeed, doses are less than 1 mSv/a in most workplaces of NORM industries with only a few exceptions in cases such as the production of uranium, thorium and rare earth elements;
- It is not always possible to predict doses;
- There is a lack of radiation protection culture in NORM industries and it is difficult to achieve the same level of culture as that existing within the nuclear industry.

The safety reports on NORM published by the IAEA address the key question: "at what level does it become necessary to regulate?" They also address the principle of the graded approach to regulation, one of the key principles on which the BSS is based. A graded approach to the radiation protection of workers is a critical component of the regulatory regime. It has a direct influence on operations and on the need to ensure that appropriate monitoring and control mechanisms are in place.

In June 2016, the Radiation Safety Standards and Waste Safety Standards Committees of the IAEA met in Vienna. A topical session on challenges in regulating NORM industries was organized, with the following objectives:

- To present the key safety issues in terms of policy, regulatory and operational aspects of NORM industries;
- To identify and prioritize areas where safety and optimization of protection is challenging;
- To suggest areas where the Secretariat could work further to enhance the internationally harmonized system.

It was generally observed that:

(a) While the need for appropriate control of NORM is widely recognized, the development of a framework for control is not emphasized as much as it should be.
(b) NORM industries cover a large variety of practices, processes and circumstances and involve multiple regulatory bodies.
(c) Occupational exposures of mine workers can be significant under certain circumstances.
(d) NORM residues cover a wide range of physical, chemical and radiological characteristics. Many have the dual characteristics of long-lived radionuclide content and the existence of bulk amounts, of the order of millions of tonnes, creating challenges for management and subsequent remediation.
(e) Many of the industry sectors concerned have limited levels of awareness, resources and experience on issues of radiation protection and the management of NORM residues (including NORM waste).
(f) There is a need for a consistent, graded and integrated approach in dealing with NORM.

There are many challenges in establishing control over NORM, of which the following should be given priority:

(1) The development of standards and guidelines for NORM that are appropriate and capable of dealing with the complexities that arise from the nature of NORM industries and from the involvement of multiple regulators and a broad range of stakeholders.
(2) The adoption of a graded approach to the regulation of NORM industries and the residues therefrom, based on good knowledge and understanding of the diverse industrial sectors involved.
(3) Synergies and system optimization with integrated consideration of radiological and non-radiological hazards.
(4) The limited experience in radiation protection that exists in many industry sectors other than uranium production.
(5) The siting and long term management of bulk amounts of NORM residues, including consideration of institutional control and financial aspects.
(6) The remediation of legacy sites and any ongoing monitoring and control of these areas after remediation.
(7) The use and recycling of NORM residues to avoid the need for long term management and eventual disposal.
(8) The engendering of stakeholder trust that enables the development of a common language for engaging stakeholders in open and transparent dialogue.

It should be noted that over the last 15 years the IAEA has developed numerous publications related to NORM industries and that the drafting of these publications has been performed with the involvement of both operators and regulatory bodies. The publications in the IAEA Safety Reports Series and TECDOC Series are geared to providing both operators and regulatory bodies (including staff at junior levels) with the necessary information for ensuring ‘good practice’ in operation and regulation.

Invited Paper

ICRP AND NORM EXPOSURE: A REPORT IN PREPARATION

(Summary)

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Task Group 76 of the International Commission on Radiological Protection (ICRP), dealing with protection against NORM exposure to individuals or contamination of the environment, was initially launched in 2007 and was relaunched in 2013. The Task Group aims to develop a report on substances with NORM that are used for other than their radioactive properties (if using for such properties then it would be considered a planned exposure situation), in industrial processes that can significantly enhance the concentration of radionuclides in products, by-products, discharges, residues and waste. Other task groups are focussing on related aspects such as contaminated sites (Task Group 98). There is also a series of reports that focus on existing exposure situations, including those involving NORM, that have been produced by ICRP Committee 4 (e.g. ICRP Publications 111, 126 and 132).

NORM arises as a result of a diverse range of practices, including mining and extractive industries, the production of coal, oil and gas, production and use of metals (such as thorium, niobium etc.), the phosphate industry and water treatment, among others. The characteristics of NORM industries are that they are already ongoing within a number of industrial sectors and tend to be large industries that are of economic importance. Furthermore, these industries tend to be subject to authorization as a result of being associated with risks from multiple hazards, but for which the radiological risk is rarely dominant and, hence, authorizations tend not to relate to radiological protection and the radiological protection culture within these industries is often poor. However, radiation doses are often higher than those associated with the classic nuclear industry, but doses are capped — there is almost no real prospect of emergency exposure situations occurring. With the culture of risk management being largely focused on non-radiological hazards, there is a need to introduce greater radiological protection to these industries, within an integrated and graded approach, ensuring that controls are commensurate with risk.

Exposures to NORM fall within the category of existing exposure situations. The source already exists, it is not deliberately introduced in an industrial process for its radioactive properties, the exposures generated are incidental, and some control is needed, commensurate with the risk. It is not so easy, however, to determine whether workers should be classified under the term ‘occupationally exposed’ since they are not truly radiation workers (the introduction of the source is unintentional). Notwithstanding, each workplace should be managed properly within a graded approach according to the dose level and the selection and implementation of protective actions.

Radiological risk at work should be integrated within a global risk management framework within industries. A reference level should be selected which, in most cases, will be less than 1 mSv/a. Higher reference levels may be appropriate (up to a few millisieverts per year) in some instances, but rarely would fall within the range of a few millisieverts per year up to 20 mSv/a.

Two series of protective actions are proposed. The first relates to workplaces and working conditions and may involve characterizing who is exposed and how, and demarcating areas and putting in place engineering controls and working procedures that are adapted to the particular situation. The second is focused on each worker individually and may involve informing, educating and training, health surveillance and the provision of personal protective equipment. The way to implement protective actions may be more or less thorough according to the situation.

For the protection of the public, the starting point is again characterization in terms of who is exposed, when and where they are exposed and how they are exposed through exposure pathway analysis and dose assessment. Reference levels should be set (below 1 mSv/a) and any required actions for public protection should be justified and optimized, with stakeholder engagement. Practically, the protection of the public is ensured through the control of discharges and waste arising or the use of residues, notably in building materials. For building materials, the materials at stake should be listed, characterized and indexed with the strategy aimed at limiting the use of materials to those with an activity index corresponding to a dose of less than 1 mSv/a.

For protection of the environment it is recommended that radiological risk is included within environmental impact assessments. This would involve radiological characterization, and the analysis of exposure pathways and of the transfer of radiological materials within the environment. Doses to non-human species should be calculated and potential effects evaluated against dosimetric criteria. Stakeholders should again be involved in the process.

A public consultation on the draft Task Group 76 report is due to take place in mid-2018, with the final report being published by the end of 2018.

STANDARDS FOR NORM INDUSTRIAL PRACTICES IN ACCORDANCE WITH THE GRADED APPROACH TO REGULATION: ELABORATION OF A STRATEGY FOR AUSTRIA

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Abstract

The Austrian Agency for Health and Food Safety was charged by the Austrian Federal Ministry for Sustainability and Tourism to prepare an expert proposal for the implementation of the NORM related sections of the European Basic Safety Standards Directive (Directive 2013/59/Euratom). This includes the identification of industrial sites associated with NORM, the definition of criteria for the various levels of the graded approach to regulation (exemption, notification, registration and licensing) according to the radiological risk, and the development of a conceptual approach to the systematic characterization of potential radiological risks at the sites concerned. The focus will be on radiologically relevant materials in practices with regard to doses received by workers and on the management of the associated residues and discharges with regard to doses received by members of the public. For some industrial activities there is a lack of information on the potential exposures; hence there will be a need for pilot studies, surveys and/or meta-analyses etc. Guidelines for industries and authorities with respect to dose assessment will also be needed.

1. INTRODUCTION

Industrial practices involving NORM are subject to the European Basic Safety Standards Directive (Directive 2013/59/Euratom) [1]. The competent authority in Austria (the Austrian Federal Ministry for Sustainability and Tourism) has charged the Austrian Agency for Health and Food Safety with the preparation of an expert proposal detailing options for the transposition and practicable implementation of the NORM related sections of the Directive. This includes the identification of practices where NORM is involved (plus the secondary industries) in Austria according to Annex VI of the Directive. Furthermore, the proposal will include the classification of practices according to their radiological risk, in accordance with the various levels of the graded approach to regulatory control (exemption, notification, registration and licensing (Chapter V, Section 2 of the Directive). An important issue will be the development of a conceptual approach to the systematic characterization of potential radiological risks due to NORM at industrial sites.

2. GRADED APPROACH FOR NORM IN AUSTRIA

In line with the graded approach, the focus will be on radiologically relevant materials in practices with regard to doses received by workers and on the associated residues (including their recycling, use as by-products, or disposal) and discharges with regard to doses received by members of the public. Figure 1 illustrates the intended graded approach for Austria. Regarding authorization, few cases are expected to fall into this level of the graded approach. As a consequence, only one category of authorization (licensing, and not registration) will be implemented for practices involving NORM in Austria.

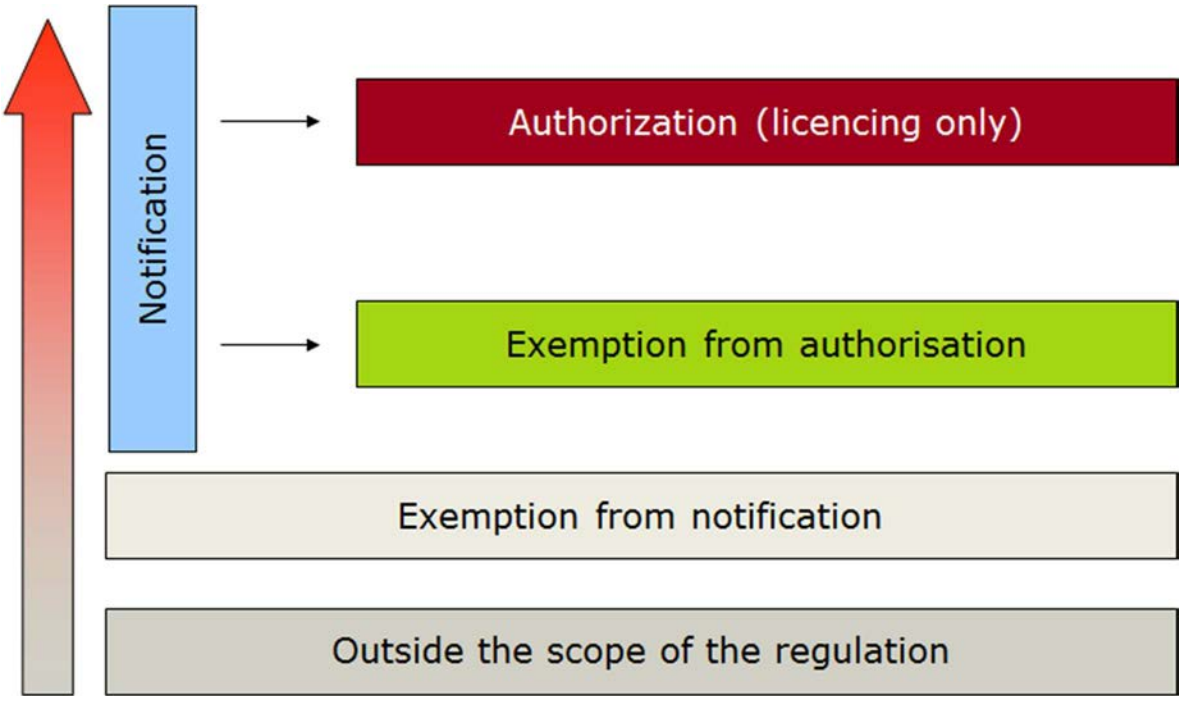

FIG. 1. The intended system of regulation for practices involving NORM in Austria, based on the graded approach (adapted from Ref [2]).

The criteria for classification according to the graded approach will be defined in an ordinance. These criteria must apply to workplaces, residues and discharges. In this respect, the Austrian standards for dose assessment will have to be adjusted.

3. ADAPTION OF THE AUSTRIAN ‘POSITIVE LIST’

For the identification of practices where NORM may occur, reports, peer reviewed literature, IAEA publications and experience from current regulatory practice were taken into account. During these searches, the focus was on the following:

- The relevant materials involved;
- Their expected activity concentrations;
- Their maximum activity concentrations;
- The contamination scenarios expected for the practices concerned;
- The disposal, recycling or by-product use of residues.

Based on these sources of information, the Austrian ‘positive list’ in the Natural Radiation Sources Ordinance was examined and will be slightly adapted to cater for the implementation of the Directive. The currently suggested version of the positive list includes two sub-sections for NORM:

(1) Practices involving possibly increased exposure due to NORM (excluding radon);
(2) Practices in which residues with increased content of uranium and thorium and their decay progeny occur.

For some practices involving NORM, there is a lack of information about the potential exposure of workers. Hence there will be a need for pilot studies, surveys and/or meta-analyses for industrial activities such as geothermal energy production, maintenance of boilers in coal fired power plants and groundwater treatment. There will also be a need for the formulation of guidelines for industries and authorities with respect to dose assessment within the notification process, in order to enable the practicable application of the graded approach.

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UPDATES TO TENORM MANAGEMENT IN THE UNITED STATES[1]

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Abstract

Management of TENORM in the United States of America is primarily left to the states to address. While based on federal statutes, states have chosen a variety of regulatory approaches to management and disposal of TENORM. Some of the states have chosen to address it on a case by case basis, whereas other states have formal programmes. Stakeholders, including members of the public and affected industries, are asking for greater clarity on the basic standards of care for management and disposal of TENORM. This paper focuses on TENORM associated with the oil and gas industry, whose practices in recent years have prompted some states to update their regulations. Examples of pending studies and legal actions will also be discussed.

1. INTRODUCTION

Regulation of NORM (or TENORM, as it is known in the United States of America) is an evolving situation. The International Commission on Radiological Protection (ICRP), in its most recent overarching recommendations (ICRP Publication 103 [1]), based its approach on exposure situations (i.e. emergency, planned and existing) rather than on processes, as previously (i.e. practices and interventions), for interpreting the principles of protection and optimization, subject to constraints. ICRP Publication 103 has led to a more prominent inclusion of natural sources of radiation in the scope of regulations and, in fact, a more aggressive approach to controlling radon exposures [2]. Updates to recommendations from the US National Council on Radiation Protection and Measurements (NCRP) are in progress [3].

In the United States of America., the primary federal agency responsible for protecting the public and environment from naturally occurring radioactivity is the US Environmental Protection Agency (EPA). The EPA's radiation protection standards, which are reflected in governmental regulations and guidance, are derived from governing federal laws controlling radiation exposures to members of the public and the disposal of the waste. Other federal agencies, such as the US Nuclear Regulatory Commission (NRC), the Occupational Safety and Health Administration (OSHA), the US Department of Transportation (DOT), and many others, have used their separate authorizing statutes to develop their own radiation standards, guidance and regulations concerning the occupational health of workers, transportation and disposal of radioactive waste and contaminated equipment, and work practices on federal lands. NORM or TENORM sometimes are contemplated specifically by these laws, whereas in other cases the law applies to materials and wastes associated with the nuclear fuel cycle, and not to NORM or TENORM specifically.

However, no federal agency has statutory authority to address TENORM in a comprehensive and holistic manner. Therefore, management of TENORM exposure and disposal in the United States of America is primarily left to the states to address. The overall existing regulatory, policy and guidance framework created by a combination of federal, state, governmental, non-governmental and industry organizations varies by industry, state and entity with respect to radiation protection and radioactive waste management. Thus, the existing

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[1] The views expressed in this paper are those of the author and do not necessarily reflect the views or policies of the US Environmental Protection Agency.

framework, while flexible to meet individual state needs and priorities, can also result in states using different criteria for radiation protection and NORM or TENORM waste disposal (e.g. existing standards for uranium process tailings or other legacy standards). States' efforts to benchmark against or achieve consistency with their peers are also complicated by their respective histories, available resources and political climates. While these criteria are derived in response to each state's unique situation (e.g. which industries and waste need to be addressed), the different approaches can cause industry and members of the public to question whether their state is being sufficiently protective or, conversely, over-restrictive of economically important industries. Industries often find it challenging to implement their programmes to address different requirements when they do business in multiple states.

However, because NORM or TENORM affect a variety of industries, with a wide range of exposure scenarios and waste profiles, it is difficult to define a single standard appropriate for all industrial sectors and exposure situations. States looking to establish standards applicable to TENORM are finding a wide variety of approaches, guidelines and benchmarks from advisory bodies and federal agencies. Some of the states have chosen to address it on a case by case basis, whereas other states have formal programmes. Industry waste management practices have resulted in some states prohibiting or further restricting certain types of discharge or disposal. In such cases, an unintended consequence can be the movement of waste to states that do not have such prohibitions or restrictions. When new information or different practices prompt states to review existing standards and rules, it is also possible for states to conclude that less restrictive conditions will provide the desired level of environmental and public health protection.

Most recently, state regulations that directly or indirectly apply to TENORM are in varying stages of development among states affected by increased activity in the oil and gas industry. Stakeholders, including industry and the public, are asking for greater clarity on basic standards of care for management and disposal of TENORM [4]. Some organizations, both governmental and non-governmental, are developing standards of good practice that may be applicable to this industry with respect to the management of TENORM. Ongoing studies are starting to provide data to support a more uniform approach, and there have been legal actions brought that may speed up the regulatory process.

2. DEFINITIONS

There are different definitions of NORM and TENORM across the US regulatory arena. For the purposes of this discussion, the following EPA definitions are provided:

- NORM: "Materials which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as radium, uranium, thorium, potassium, and their radioactive decay products, that are undisturbed as a result of human activities" [5]. Radiation levels presented by NORM are generally referred to as a component of "natural background radiation."
- TENORM: "Naturally occurring radioactive materials that have been concentrated or exposed to the accessible environment as a result of human activities such as manufacturing, mineral extraction, or water processing. Technologically enhanced means that the radiological, physical, and chemical properties of the radioactive material have been altered by having been processed, or beneficiated, or disturbed in a way that increases the potential for human and/or environmental exposures" [5].

The definition of TENORM put forward by the Conference of Radiation Control Program Directors (CRCPD) [6], which is used by many states, is limited to the increased concentration of radioactivity in materials and does not amplify the need to include materials that have not been modified by human activities, yet have been disturbed in such ways that they can present an additional potential for exposure. This basic inconsistency in definitions has resulted in differing management and regulation of some NORM and TENORM in different states. The CRCPD has created a working group to address this definitional question.

3. DISCUSSION

The EPA is authorized under a number of environmental laws to prepare regulations or guidance to limit radiation exposures of the public from certain sources of radiation, or from practices that can expose members of the public to radiation. As described in a 1999 National Academy of Sciences report [7], these include[2]:

- Operations of uranium fuel cycle facilities (Atomic Energy Act (AEA));
- Radioactivity in drinking water (Safe Drinking Water Act of 1974 (SDWA));
- Radioactivity in liquid discharges (Clean Water Act of 1977 (CWA));
- Uranium and thorium mill tailings (Uranium Mill Tailings Radiation Control Act of 1978 (UMTRCA); AEA);
- Radioactive waste management and disposal (AEA);
- Remediation of radioactively contaminated sites (Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA); AEA);
- Airborne emissions of radionuclides (Clean Air Act (CAA));
- Indoor radon (Indoor Radon Abatement Act of 1988);
- Naturally occurring and accelerator produced radioactive materials, including TENORM, which are not subject to regulation under the AEA (Toxic Substances Control Act of 1976 (TSCA)).

Many of these programmes are implemented at least partially by the states, typically through authority delegated by the federal agency.[3] State regulations for the control of TENORM can differ from one state to another in numerous ways, depending on the presence in a state of industries that generate such waste or, in some cases, whether a state has a requirement to adopt and incorporate regulations that were developed by the CRCPD (e.g. Colorado). States also differ in how they distribute regulatory responsibility for TENORM management. These differences among states that have TENORM regulations fall into three categories:

(i) States that have their radiation protection and radioactive waste disposal requirements residing within a department of health or radiation control agency;
(ii) States whose radiation protection and waste disposal regulations reside within a department of natural resources or environmental protection (or similar agency);
(iii) States whose radiation protection and waste disposal regulations for TENORM have been developed by a state oil and gas conservation commission or similar agency.

Some states fall into more than one of these categories. There are also some states that have not developed their own TENORM regulations to date. Some differences between states,

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[2] This abbreviated list is cited directly from NAS [7], but that list does not include some named amendments to those acts. As an example, CERCLA was amended by the Superfund Amendments and Reauthorization Act (42 U.S.C. 9601 et seq.) of 1986.

[3] The Resource Conservation and Recovery Act (RCRA) is another federal statute that is important for the management of TENORM wastes. RCRA applies to the management of solid and hazardous waste. While it does not address radioactivity, disposal facilities permitted under RCRA are used for disposal of certain TENORM wastes. Nearly all states have permitting authority delegated from the EPA for hazardous waste management. Solid waste management does not require delegation, but is solely the purview of the states.

or even within state programmes, may arise from the approach taken to the issue by the regulatory authority, for example addressing TENORM from a waste management perspective as opposed to a radiation protection perspective.

Lacking a definitive, complete set of federal standards for TENORM to follow, states have elected to be flexible to meet their own needs in choosing whether or not to develop a TENORM standard, and what to include in it. This is understandable in that, over the decades, different states have faced challenges from different industries that have predicated some action (e.g. phosphogypsum, drinking water treatment, hard rock mine tailings, monazite sands and rare earths). Some states have solely chosen to adopt NRC radiation protection standards as part of their Agreement State[4] roles, and then have added EPA requirements for water protection and waste disposal, as well as (possibly) OSHA/DOT occupational worker protections.

4. UPDATE ON TENORM FROM OIL AND GAS PRODUCTION

In addition to other ongoing industries affected by TENORM mentioned above, the increase in activities associated with the oil and gas industry is the most recent challenge that states have to address with respect to TENORM. A number of major oil and gas producing states that knew about TENORM contamination within their borders established TENORM regulations as early as the 1990s, such as Louisiana [8], Texas [19] and New Mexico [10]; those regulations may or may not be consistent with the most recent federal, state and non-governmental organization standards. Others have followed guidance developed by non-governmental organizations such as the Interstate Oil and Gas Conservation Commission (IOGCC) or CRCPD, and some have added selected TENORM standards to existing public and occupational radiation protection standards, whether they are Agreement States or not.

In its own survey of states (not all having oil and gas production), the Association of State and Territorial Solid Waste Management Officials (ASTSWMO) [11] found that 27 of 38 states that answered the corresponding question provide regulatory information for the licensing of accelerator produced radioactive matieral (NARM) or naturally occuring radioactive material (NORM or TENORM) in the state. Of those, six did not have regulations specifically for TENORM, but NARM, NORM or TENORM were licensed under the state authority for regulation of radioactive material. Of the 40 states that responded to another question on whether they had regulations governing disposal of NARM or TENORM, 22 said they did. At the time, only three states had adopted the CRCPD Part N Suggested State Radiation Control Regulation, and a fourth (Alabama) responded that it had used the radiological limits in Part N. With respect to the disposal of TENORM in municipal, industrial or hazardous waste landfills, 34 states responded, and 16 indicated that they do allow such disposal while only 2 of those did not allow it in such landfills. It is apparent from the ASTSWMO survey that there is a great divergence in state approaches to navigating TENORM regulation and especially in using solid waste management facilities for the disposal of TENORM wastes.

The Health Physics Society and American National Standards Institute TENORM standard [12] recommended a public dose limit for all combined controllable sources of radiation of less than or equal to 1 mSv per year (100 mrem/yr), which would mean a smaller dose from any individual source of TENORM. Yet the ASTSWMO survey found that nearly all of the states whose regulations were reviewed continue to use a 1 mSv (100 mrem) annual standard for exposure limits that would not generally partition the annual exposures to oil and gas TENORM below 1 mSv (100 mrem).

EPA's UMTRCA standards establishing a radium concentration limit of 0.185 Bq/g (5 pCi/g) in contaminated soil surface have been generally adopted by oil and gas producing states, but up to 1.11 Bq/g (30 pCi/g) in some cases, including landspreading, has been allowed

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[4] Section 274 of the AEA provides a basis for NRC to enter into agreements to relinquish to states portions of its regulatory authorities. Such states are known as "Agreement States." There are 37 Agreement States.

in Louisiana and Texas, and included in the IOGCC (2001) standard. As noted in the next paragraph, some states have applied the UMTRCA cleanup standard in non-cleanup contexts.

Public awareness of TENORM, and most recently oil and gas TENORM, has often been prompted by news reports related to large quantities of TENORM waste and discharges that do not fit into a state's existing regulatory framework. This tends to create an impetus to address the issue for the regulatory agencies, the industry looking for the regulatory framework to work within, and the public that is uncertain how to interpret unanticipated waste management and exposure situations. For example, based on limited samples of residues containing TENORM provided by industry to the state environment department, North Dakota used recommendations from Argonne National Laboratory [13] to develop its waste disposal standards. Previously, the patchwork nature of state regulatory frameworks may have resulted in possibly illegal disposal in North Dakota [14], with some states like Ohio and West Virginia also unexpectedly receiving TENORM waste [15]. Ohio subsequently adopted the UMTRCA cleanup standard as an exemption limit for TENORM. Materials above the exemption limit cannot be disposed of in solid waste landfills in Ohio [16].[5] In another recent case, TENORM waste from Ohio, Pennsylvania and West Virginia may have been disposed of illegally in Kentucky [17], which is still developing its own standards and regulations.

Some states have applied regulatory approaches to TENORM waste from other industries that could prove applicable to oil and gas residues. In Illinois, persons producing or possessing residues or sludge resulting from the treatment of water or sewage and containing naturally occurring radium from groundwater are exempt from licensing provided they contain a combined radium (^{226}Ra and ^{228}Ra) concentration less than or equal to 7.4 Bq/g (200 pCi/g) (dry weight basis). Ultimately, the exemption came about in response to petitions for higher land application rates for radium. The state also considered its experience with response efforts at landfills and scrap yards handling treatment residues and equipment contaminated with radium, the need to create awareness and the agency's responsibility to protect the health and safety of municipal workers. Specific details on requirements for land application, field sampling, disposal and annual reporting were codified in the resulting rule. While those entities producing these residues as a result of the treatment of drinking water or wastewater are exempted from licensing, there are specific sampling, reporting and disposal conditions that now exist [18].

States have taken similarly diverse approaches to managing liquid effluents from oil and gas activities, in particular produced water. Not all states permit discharges of produced water containing TENORM into their streams, but some do, with the potential for radium concentrating in stream sediments and water potentially being used as source water for community water systems (CWS). Pennsylvania has identified some areas of contamination at discharge outfalls [19].

Given the potential for future cleanup of TENORM from spills and improper disposition of wastewater, sludges and solids, it is important to consider what the states will use to derive the action levels for response; possibilities include the EPA risk range for CERCLA (Superfund) and the exemption level adopted by many states for radium, e.g. 0.185 Bq/g (5 pCi/g). It is possible that states may use a variety of criteria for different situations.

There is a paucity of publicly available data relative to TENORM associated with more recent oil and gas development practices. A recent investigation by Pennsylvania is the most current study on TENORM associated with natural gas in the Marcellus formation [19]. EPA is conducting a study looking at impacts on drinking water from unconventional oil and gas that includes some discussion of TENORM [20]. As part of its broader TENORM programme,

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[5] Ohio has adopted the CRCPD definition of TENORM, so this landfill restriction does not apply to waste streams, such as drill cuttings, where radionuclide concentrations have not been increased. Such waste streams are considered NORM.

EPA has also evaluated TENORM in uranium overburden [21, 22] and in the copper belt in southwest Arizona [23].

Legal challenges relating to TENORM have resulted in settlements, many taking decades to work through the appeals process. These include cases involving pipe rattling yards in Louisiana that resulted in surface contamination and worker exposure and the Martha Oilfield in Kentucky that spread contamination over the surface from liquid discharges [24]. Many communities are trying to restrict where drilling can be utilized, but most of those actions do not specifically cite TENORM. There has been a recent suit brought by the National Resources Defense Council and others against the EPA for allegedly not updating solid waste disposal regulations in a timely manner [25]. TENORM is specifically called out in the complaint.

5. CONCLUSION

In the United States of America, the primary federal agency responsible for protection of the public and environment from naturally occurring radioactivity is the US Environmental Protection Agency. NORM and TENORM management in the United States of America is primarily left to the states that implement a variety of federal statutes and regulations, as well as developing their own where federal regulations do not exist. Some states have updated their regulations to address increased waste from oil and gas exploration and production. Market forces play a role in the disposal of TENORM residues from a variety of industries, but disposal is currently being impacted by the increase in TENORM waste from oil and gas expoloration and production. There is a paucity of publicly available data relative to TENORM from these more recent drilling practices that makes comprehensive evaluation of its characteristics, occurrence and management difficult at this time.

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Invited Paper

CHALLENGES IN IMPLEMENTING THE CHANGE IN REGULATIONS IN THE NORM INDUSTRY: THE NEW EUROPEAN DIRECTIVE

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Abstract

Advances in radiation protection with regard to NORM have become evident since 1996, acquiring a worldwide dimension that did not exist 20 years ago. New regulations for the control of exposure to NORM have become established and knowledge on exposure levels has clearly improved, diminishing the controversial aspects associated with radiation protection in NORM industries. Nevertheless, not all the controversial aspects have disappeared, there being a perception in the community involved in the NORM issue that the treatment of exposure to natural sources in international standards has been unnecessarily complicated and confusing, giving rise to misunderstandings and differences in their implementation, mainly in basic and essential concepts. The new European Council Directive on radiation protection, which replaced the 1996 version, was published only quite recently. The new Directive has a simplified structure and its revision has met one of the main objectives, namely to improve the treatment of exposure to natural sources. This has been mainly achieved through the greater use of quantitative rather than qualitative criteria. It seems evident that the new Directive will provide greater clarity on the control of exposure to natural sources, although its application will be far from being a trivial task, there being several unavoidable challenges remaining for proper implementation in the NORM industry. The most important challenges in this implementation, as seen by the author, are discussed in this paper.

1. INTRODUCTION

It was about 20 years ago when some countries, mostly European, started to introduce measures to regulate exposures arising from a wide range of natural sources, in particular from minerals other than those associated with the extraction of uranium. Two important milestones in this regard were reached in 1996, with the publication of the IAEA International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources [1] and the European Council Directive 96/29/Euratom laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation [2]. But only one year after the publication of the European Directive, the first international NORM Conference was held in Amsterdam specifically in response to the concern generated in the European chemical industry about the implications of the implementation of these regulations. Its novelty, the lack of experience of all the parties involved and also the confusing treatment of some key points in the international standards were the cause of a certain degree of chaos during the first few years. It was the time where misunderstandings in the interpretation of several requirements of the legislation were not unusual, and where a tendency for overregulation was evident.

2. NEW RADIATION PROTECTION STANDARDS

Since 1996, new regulations for the control of exposure to NORM have become established and the knowledge about levels of exposure has clearly improved. But a perception within the community involved in the NORM issue, albeit to a decreasing extent with the progression of time, is that the treatment of exposure to natural sources in international standards has been unnecessarily complicated and confusing, giving rise to misunderstandings and differences in the interpretation of the standards, mainly in basic and essential concepts.

New International Basic Safety Standards were published quite recently by the IAEA [3] and have been implemented within the European Union through a new European Council Directive [4]. The requirements set out in the new Directive are in line with the 2007 Recommendations of the ICRP [5]. In particular, the three types of exposure situations specified by the ICRP (planned exposure situations, existing exposure situations and emergency exposure situations) are reflected in the structure of the new standards. 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 in the past, is now based on the use of so-called reference levels. Only a few exposures to natural sources are, by exception, subject to the requirements for planned exposure situations. The main example in this regard is exposure to material (other than commodities such as food, drinking water, fertilizer and construction material) with a radionuclide concentration exceeding 1 Bq/g for the U and Th decay series or 10 Bq/g in the case of ^{40}K — in other words, exposure to NORM. In addition, for the first time, numerical criteria for exemption and clearance of NORM have been included, with exemption being determined on the basis of dose commensurate with the natural background (about 1 mSv/a) and clearance being determined on the basis of activity concentration (1 Bq/g for U and Th series radionuclides and 10 Bq/g for ^{40}K). Also, as in the previous standards, exposures that are unamenable to control are excluded from the scope of the new standards. This is particularly important in the case of natural sources, because it leads to the exclusion of exposures such as to ^{40}K in the body, cosmic radiation at the surface of the earth and exposure associated with undisturbed terrestrial areas.

More specifically, the requirements for existing exposure situations should be applied to the following exposures to natural sources: exposure to radon in homes and in most workplaces, exposure to commodities (including food, feed, drinking water, agricultural fertilizer and soil amendments, and construction material) irrespective of the activity concentrations, exposure to residual radioactive material in the environment irrespective of the activity concentration, exposure from past activities that were not regulated and exposure to any other material provided that its radionuclide activity concentration does not exceed 1 Bq/g for each radionuclide of the ^{238}U and ^{232}Th series and 10 Bq/g for ^{40}K.

Only exceptionally are there some human activities where exposure to natural sources should be controlled in accordance with the requirements for planned exposure situations. They are the following: (a) Exposure to material — other than food, 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 (such material being defined as NORM); (b) public exposure delivered by effluent discharges or the disposal of radioactive waste arising from the facilities concerned, irrespective of their activity concentrations; (c) Occupational exposure to radon, irrespective of the concentration, in situations where the exposure to other U and Th series radionuclides is controlled (as a planned situation); (d) Occupational exposure to radon in situations where the concentrations remain above the reference level after remedial action has been carried out.

3. CHALLENGES FOR REGULATION OF NORM

It seems evident that the new European Directive will provide greater clarity on the control of exposure to natural sources although its application will be far from being a trivial task, with several unavoidable remaining challenges for proper implementation in the NORM industry. Such challenges include the following:

(1) The requirements in the new Directive for existing exposure situations include the concept 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. The reference level approach replaces the 'action level' approach in the previous Directive — action levels were defined as the levels of dose rate or activity concentration at or below which remedial action or optimization was not normally necessary. With the new Directive, some effort should be made for the proper implementation and full understanding of this 'reference level' approach. The tendency observed in the first months of implementation of the new Directive to simply interchange the concepts of action levels and reference levels (thinking that the change is merely one of terminology) should be properly corrected. The 'reference levels' sometimes have been used as limits, defeating the purpose of optimization.

(2) It is clear that the control of exposure to NORM in industrial activities should be performed in accordance with the requirements for planned exposure situations, in the same way as the control of public exposures due to release or disposal of wastes generated in NORM industries. This control must fulfil one of the key principles in the Directive — it should be commensurate with the characteristics of the practice or the source and with the magnitude and likelihood of exposures. In the case of NORM industries, this is particularly relevant because the exposures are generally (but not always) moderate with little or no likelihood of extreme radiological consequences from accidents. This philosophy of the graded approach to regulation is maintained unchanged in the new Directive, following the principle of optimization of protection. The only important difference from the previous Directive is that numerical criteria have been introduced for the basic concepts of exemption and clearance. In particular, where a radionuclide has an incidental presence, and where bulk quantities of material may be involved (as in many NORM activities), the new Directive makes provision for exemption using a dose criterion of about 1 mSv/a commensurate with natural background levels, as well as for clearance using criteria expressed as activity concentrations not exceeding 1 Bq/g for uranium and thorium series radionuclides and 10 Bq/g for ^{40}K. In this sense, a continuous didactic dissemination of the basic concepts of exclusion, exemption and clearance is needed, because they are being applied confusingly or wrongly by a non-negligible fraction of the parties involved in radiation protection for NORM.

(3) In some cases, there are situations where doubts can appear about which type of requirements (for planned or existing exposure situations) should be implemented. By default these cases should be treated as existing exposure situations and only in exceptional cases as planned exposure situations, following the philosophy of the European Directive that treats the great majority of exposures to natural sources as existing exposure situations.

(4) Obviously, although standards have been developed for the NORM industry in general without exceptions, it is clear that individual NORM industries are very different as are the practical radiation protection challenges they face. No single approach is appropriate for all NORM industries or for all industrial processes. The idea of a common protocol to control the exposures in all the NORM industries, i.e. a uniform approach to regulation of NORM, should be disregarded in favour of an industry specific approach. The importance of this point is reflected in the publication of the 'positive list' of industries proposed for Europe and (in the IAEA industry-specific publications in the Safety Reports Series) worldwide.

(5) Through the individual studies performed in NORM industries it is clear that, in most of the workplaces of NORM industries, worker doses are less than 1 mSv/a and the majority of studies that have been published or presented in various international conferences have concentrated on the determination of individual doses in order to establish whether there is a need for regulatory control. Only in a very limited number of cases has information been provided on how such doses might then be optimized in practice. Additional detailed studies should be performed in NORM workplaces where optimization of doses is needed, devoting special emphasis to the evaluation of possible alternative approaches to decrease the occupational doses in practice.

(6) On the other hand, it is interesting to note that methodologies for the realistic assessment of worker doses suffer from non-standardized approaches. There is therefore a need to plan how such standardization can be achieved. The emphasis should be on actual monitoring data (individual and workplace) to ensure that the dose estimates are realistic, rejecting the approaches based on theoretical models.

(7) A key issue associated with the application of the new regulations in NORM activities is the legacy issue. Many NORM related industrial sites were abandoned in the past with little or no remediation and inefficient residue management. Such sites may include tailings facilities, fertilizer plants, thorium gas mantle factories, metal refineries and old oil production fields. Many of these legacy sites, including those in the vicinity of urban areas, now need remediation. In terms of the European Directive, the requirements for existing exposure scenarios must be applied. This poses the challenge of establishing coordinated international efforts for remediation. The goal of the remedial actions must be the timely and progressive reduction of the hazard and eventually, if possible, the removal without restrictions of regulatory control from the area. The nature and extent of remedial actions must be commensurate with the risks associated with the existing exposure, and must be justified in the sense that they do more good than harm. The form, scale and duration of the remedial actions must be optimized so as to produce the maximum net benefit and must be selected from a set of justified options for remediation. The optimized protection actions must be implemented with the objective of reducing doses to below the reference level. However, in terms of the new reference level approach, this is no longer sufficient. Exposures below the reference level must not be ignored and possible reductions in such exposures need to be investigated to ensure that the remedial actions are optimized.

(8) Another important issue in NORM industries is the management of NORM residues (including those designated as NORM waste). This management is not a simple task, since such residues can be generated in a variety of forms, including solids in small quantities (scale, sludge, tailings, slag), solids in large quantities (mine tailings, waste rock piles, by-product piles), liquids (spent process water, spent solvents) or gases (from high temperature processes). NORM residues should be managed in accordance with the basic principles embodied in the European Directive. For some NORM industries, especially those producing low or medium volume residues with higher activity concentrations, disposal as waste in a suitable repository may be the only solution. In recent years, it is becoming increasingly accepted that these NORM residues may be disposed of in a manner similar to that for other hazardous wastes, rather than in facilities designed for the disposal of high activity radioactive waste containing artificial radionuclides. However, the problem is still the lack of suitable facilities that are prepared to accept NORM waste.

(9) For residues with moderate concentrations of radionuclides, there is increasing acceptance of recycling or use of such residues as by-products, rather than disposal as waste. The opportunity for recycling or by-product use of a NORM residue depends on a variety of factors such as the type of residue, the rate and place of generation and the local

market conditions, taking into account factors such as acceptability to the public, the existence or otherwise of uniform approaches to recycling and concerns about radon exposure in the case of building materials. For that reason, the recycling of these residues should be based on prior detailed assessments based on experimental studies to determine the potential health, environmental and radiological impacts of the product generated, as well as on assurances that the recycled residue will have the necessary characteristics and behaviour for the application in question.

4. CONCLUSIONS

The number and magnitude of the challenges summarized in the paragraphs above can be considered as quite important, but the writer, having been involved in NORM for more than 20 years, wishes to conclude by indicating that the actual situation of radiation protection in the NORM industry can be regarded with optimism. The experience gained in the field has been considerable and the regulations are becoming increasingly clear and properly applied. The challenges described in this paper are those that remain after having taken a good number of steps in the right direction. We are fortunate enough to be far from the initial erratic and chaotic situation that existed with respect to the application of NORM regulations some years ago, and are approaching the desired degree of maturity where radiation protection is a well established subject that can be handled routinely by NORM industries.

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Poster Presentation

RADIOLOGICAL RISK CLASSIFICATION OF NORM INDUSTRIES IN BRAZIL

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Abstract

This work is based on the identification and recognition of NORM industries in Brazil. The main NORM industries are found in mining, energy and fertilizer production. The handling of radioactive material and disposal of waste from these NORM industries is in accordance with the regulations of the National Commission on Nuclear Energy and the safety requirements of the IAEA. In this work, the classification of radiological risk does not address nuclear power production plants or even oil production, thereby restricting the industries to mining and fertilizer production. After surveying the industries, a radiological risk classification was performed based on the activity concentration of each radionuclide, taking into account the current regulations. The results show, for various regions of Brazil, a wide range of NORM industries having a potential radiological impact on workers, members of the public and the environment.

1. INTRODUCTION

Some industrial activities, especially those related to minerals extraction, are associated with NORM, either as a main activity or as part of the industrial process. All of them show many problems during handling. These radiological problems have three types of impact: occupational exposure, public exposure and environmental exposure. The occupational exposure is related to the radiological risk due to the close contact with NORM that could be from a few hours to long periods of the day depending on the industrial activity. Public exposure is related to NORM waste management, treatment and disposal. An environmental exposure impact occurs when, for example, the industry needs to process water in contact with NORM, with possible consequences due to contamination of the marine fauna.

In terms of the IAEA's international standards, NORM is radioactive material containing no significant amounts of radionuclides other than radionuclides of natural origin [1]. These radionuclides (^{238}U, ^{235}U, and ^{232}Th decay series and ^{40}K) are the primordial radionuclides found in rocks and soil and in materials in which the radionuclide concentrations have been elevated as a result of processing. In the industrial production process, NORM can be found in the mining of metals, the phosphate industry, coal mining and power generation from coal, oil extraction, the rare earths and titanium dioxide industry, the zirconium and ceramics industries and water treatment [2]. The Brazilian regulatory body — the National Commission on Nuclear Energy (CNEN) — has promulgated a national regulation that establishes safety and radiation protection requirements for mining and industrial facilities that handle, process and store minerals, raw materials, waste rock, slag and other waste containing radionuclides of natural origin, simultaneously or separately and which can at any time during or after the operational phase cause undue radiation exposure of workers and members of the public [3]. The regulation also defines “mining–industrial installation” as an installation with uranium and/or thorium associated or an installation where raw materials containing radionuclides of natural origin (uranium and/or thorium series) are mined, processed and industrialized, including waste storage. The regulation classifies NORM facilities into three categories:

Category I:	Facilities with a radionuclide activity concentration in solid material exceeding 500 Bq/g;
Category II:	Facilities with a radionuclide activity concentration in solid material exceeding 10 Bq/g but not exceeding 500 Bq/g;
Category III:	Facilities with a radionuclide activity concentration in solid material not exceeding 10 Bq/g giving rise to an annual dose to workers exceeding 1 mSv above background levels.

2. METHOD

To carry out the radiological risk classification, it was necessary to use a benchmark. The parameters used were the activity concentrations (in becquerels per gram) of ^{238}U and ^{232}Th present in the material. As the activity concentrations of ^{238}U and ^{232}Th ranged from about 0.2 to 810 Bq/g and these values are directly proportional to radiation dose, a classification of radiological risk into low, medium and high risk was done. Another very important consideration in this classification was the harmonization with the national NORM regulation [3], which uses the same quantity for the classification of facilities. The proposed radiological risk classification was 0.1–1 Bq/g for low risk, 1–10 Bq/g for medium risk and 10–100 Bq/g for high risk.

3. RESULTS

Nineteen NORM industries in five Brazilian regions were surveyed and assigned an appropriate risk classification. The results are shown in Table 1 and Fig. 1. Nine industries were classified as low risk, four as medium risk and six as high risk.

TABLE 1. RISK CLASSIFICATION OF 19 BRAZILIAN NORM INDUSTRIES

State	Company name	Product	Activity concentration range (Bq/g)	Risk classification
Northern region:				
AM	Pitinga	Cassiterite, iron, niobium	10–100	High
PA	CVRD–Serra do Sossego	Copper concentrate	0.1–1	Low
PA	CVRD Projeto 118	Copper concentrate	1–10	Medium
North-eastern region:				
RN	Borborema	Uranium, thorium	0.1–1	Low
PB	Borborema	Uranium, thorium	0.1–1	Low
PB	Mataraca	Titanium	0.1–1	Low
BA	Camaçari	Titanium	0.1–1	Low
Central western region:				
GO	Catalão	Iron, niobium	10–100	High
South-eastern region:				
MG	Araçuaí	Niobium	0.1–1	Low
MG	Araxá	Niobium	10–100	High
MG	Nazareno	Tantalum, niobium	10–100	High
MG	São Tiago	Tantalum, niobium	10–100	High
MG	Araxá	Beneficiated phosphate rock	1–10	Medium
MG	São João del Rey	Tantalum, niobium	10–100	High
MG	São João del Rey	Manganese	0.1–1	Low
MG	Tapira	Beneficiated phosphate rock	1–10	Medium
SP	Pirapora do Bom Jesus	Tin	0.1–1	Low
SP	Cubatão	N–P fertilizer	1–10	Medium
Southern region:				
PR	Figueira-Cambuí	Coal	0.1–1	Low

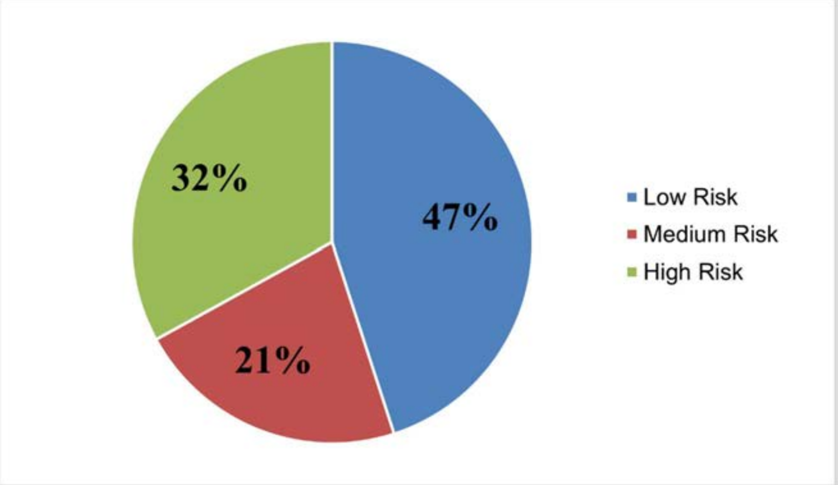

FIG. 1. Risk classification of 19 Brazilian NORM industries.

4. CONCLUSIONS

This survey of the main NORM industries in Brazil provides an overview of the types of mining activity in each region and the associated radionuclide activity concentrations. The south-eastern region of Brazil stands out with the largest number of NORM industries and also with the largest number classified as high risk. This radiological risk classification provides a broad view of NORM industries in the various regions with a potential radiological impact on occupational, public and environmental exposures and can be used to identify the need for further measurements and possible radiation protection measures.

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RISK COMMUNICATION (WORKERS AND PUBLIC)

(Session 2)

Chairperson

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Brazil

Invited Paper

MISSION (IM)POSSIBLE: COMMUNICATING ABOUT NORM TOPICS AND STAKEHOLDER ENGAGEMENT

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Abstract

This presentation gives an overview of different constraints related to communication about NORM, highlighting common stakeholder involvement failures in different remediation processes involving NORM. It presents case studies and solutions derived from lessons learned from NORM contamination in Europe in general and from mixed contamination (chemical and radiological) in Belgium in particular. Our purpose is to demonstrate the need to openly and transparently communicate with the public about radioactivity, to involve stakeholders in decisions related to NORM and to show that risk communication and stakeholder involvement are not only “mission possible”, but also “mission unavoidable”.

1. INTRODUCTION

Small scale and large scale environmental remediation programmes, as well as decommissioning of NORM installations, have been implemented in many European countries in order to reduce prolonged radiation exposure to NORM. In 2016 the following nine sites in Belgium were registered for the disposal of waste types that included NORM waste [1]:

- Four surface repositories for waste from individual facilities in the phosphate industry (phosphogypsum stacks);
- One surface repository for waste from a titanium dioxide production facility;
- Two surface repositories for hazardous waste;
- One surface repository for non-hazardous waste;
- One incinerator for hazardous waste.

Although the last four sites in this list were able to accept waste from any Belgian source, and were thus not designated specifically for the disposal of NORM waste, they accepted 274 t of NORM waste in 2015.

2. CHALLENGES

While the scientific and technical aspects of NORM have been extensively addressed, studies on challenges due to socioethical and cultural aspects, as well as communication and stakeholder engagement aspects, are lacking. Challenges related to these aspects in remediation programmes arise mostly from the different opinions, perceptions, attitudes and concerns towards the risk and benefit of remediation programmes by stakeholders, and also from poor communication and delays in the involvement of stakeholders [2]. The main communication and stakeholder involvement challenges are that the experts and the public frequently disagree when it comes to risk assessment. Several studies related to these differences have demonstrated that experts have in general a lower perception of radiological risks than the general public [3–4]. More specifically, it was shown in Ref. [5] that the general population has a higher risk perception for nuclear waste and natural radiation than the experts. These differences can be explained by the psychometric approach [6] which suggests that familiarity with the hazard, knowledge, personal control and voluntary status decrease the risk perception [7, 8]. Those involved in communication about NORM and related remediation processes have to take into account not only radiological risks, but also changes in social attitudes (e.g. stigma), psychological effects (e.g. distress, depression, disturbance of daily life), protective measures (e.g. diet, removal of 'backyards') and economic threats (e.g. decrease of property value, boycott of products). While lay people often do not have a clear idea of the nature of radioactivity and NORM, it is expected and, in many cases, required that industry and authorities communicate and engage with stakeholders about risks related to NORM.

3. KNOWLEDGE ABOUT RADIATION

On the one hand, public communication about radiological risks and stakeholder involvement is very demanding owing to the limited knowledge of the public, different risk perceptions, and views and feelings about NORM. In Belgium, for instance, knowledge of radiation is rather poor, even after an intense communication campaign related to radon[1], medical imaging[2] and nuclear emergency response organized by authorities, and after an information campaign (the Belgian Nuclear Forum) focused mainly on nuclear applications organized by the nuclear industry. In a public opinion survey conducted by the Belgian Nuclear Research Centre in 2013 and 2015, one in three respondents thought that: "natural radioactivity is never dangerous". At the same time, one in four thought that every radioactive substance "becomes with time more and more radioactive", while half knew the correct answer to the question [9].

On the other hand, importance of knowledge is often overestimated [10]. Many risk communicators mistakenly measure the success of risk communication by what the population knows about the risk, and whether it believes it knows enough to make a decision. But knowledge may not always play a role in determining people's behaviour. Knowledge about radon, for example, is uncorrelated with actually doing a home radon test [11]. It has also been shown that people who take risks are not necessarily less knowledgeable than those who do not take risks [12]. Moreover, it has been found that general knowledge about radiation had little or no influence over lay persons' satisfaction with the information about radiation provided by different communicators in Belgium and France [9]. Quoting from Ref. [13]:

> "Confidence in authorities was revealed in Belgium and France as more important for satisfaction with information than risk perception. An additional study for the Belgian sample showed that both perceived trustworthiness and technical competence influence satisfaction with information, but their relative importance depends on the communicator".

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4. COMMUNICATION AND STAKEHOLDER ENGAGEMENT

It is confirmed by projects such as the IAEA CIDER project [14] and the European Commission Seventh Framework Programe EAGLE project[3] and Horizon 2020 ENGAGE[4] and TERRITORIES projects that open, transparent and timely communication (for instance at the early stages of a remediation project) and stakeholder involvement are beneficial for industry, authorities, lay people and other stakeholders. Stakeholder engagement has been proven effective in bridging the gaps between opinions, views and expectations. Through stakeholder involvement, public concerns can be addressed in an open and transparent way, trust can be built between the different parties and mutual understanding can be increased. Furthermore, stakeholders contribute to better processes and better outcomes, developing during the process a certain ownership of the solutions to be implemented. Sound communication and stakeholder involvement lead to effective, democratic, ethical and transparent decisions, important for NORM related governance. Yet, stakeholder involvement is a complex process that demands a lot of time and human and financial resources [2, 14].

The Belgian site at Olen serves as an example to demonstrate the importance of the stakeholder engagement process. This site is taken as a case study of societal uncertainties in remediation projects under the European Commission Horizon 2020 TERRITORIES project[5]). Quoting from Ref. [15]:

> "Mixed contamination (chemical and radiological) at the site is due to the metallurgic industry, specifically radium extraction and production of radium sources from 1922 till 1969. This activity led to significant contamination, both inside the factory premises and outside: dumping sites, the banks of a nearby river and a few streets of the neighbouring town. The first environmental study was conducted in 1960 after which the mixed pollution (heavy metals and radioactive materials) was recognised. In the 1980's the local community and green activists pressured the authorities to redo the study. In 1993 the report was made available and showed pollution in river banks, waste disposal….
>
> "Subsequent to this, a press conference was organised by the local community. A committee of 20 people, including authorities and company, was established but decisions were postponed. It was not until 2000 that a company and nuclear waste agency (ONDRAS/NIRAS) came up with the BREAM project. This resulted in 2002 in the organisation of a first public meeting, after which a brochure was published with explanation of the project and timing. Additionally, a working group was established. In 2003 an outside company was appointed for the remediation and in 2004, OVAM took over the remediation based on the idea that mixed contamination leads to mixed responsibility. In 2004, there was also an agreement reached on the financial aspects and an external communication officer was hired. In 2006 the remediation started, in 2008 the remediation finished, and in 2009 new measurements were done which showed that not all sites were remediated due to for example land instability. In Olen the stakeholders involved comprise the local population, local authorities, government institutions, industry, concerned companies, media, private actors, politicians etc…

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[3] http://eagle.sckcen.be.

[4] http://www.engage-h2020.eu/.

[5] TERRITORIES – To Enhance unceRtainties Reduction and stakeholders Involvement TOwards integrated and graded Risk management of humans and wildlife In long-lasting radiological Exposure Situations.

> "In general the public was happy that somebody explained the situation (including the risks and the remediation strategy) to them. The public was not necessarily aware of radioactivity and wondered why such dramatic interventions were needed. From this a working group with local population was established to create dialogue and consultation opportunities".

The relevance of stakeholder engagement and communication in complex decision making is recognized by international organizations [16–19]. Stakeholder involvement in decommissioning and environmental remediation programmes is also set out in different academic and legal texts, such as the European Directives 97/11/EC [20] and 2003/35/EC [21]. In addition, the Aarhus Convention on access to information, public participation in decision making and access to justice in environmental matters [22] states that the public should be involved at the earlier stages of the environmental impact assessment process, and thus also in remediation programmes [2]. Figure 1 illustrates the various groups of stakeholders and gives an idea of the decisions to be taken by stakeholders at various stages of the environmental remediation process.

The importance of early involvement of stakeholders in the remediation process is often acknowledged, but practice shows that this happens mainly at a later stage when remediation projects are jeopardized. The IAEA CIDER project [14] points out that involvement should start with listening to stakeholders' concerns and demands, agreeing on the ground rules to establish a dialogue and, if possible, jointly framing the issue. Quoting from Ref. [2]:

> "The involvement of an independent facilitator agreed by all parties and/or the international community can help to open up dialogue. In addition, providing stakeholders with financial resources to choose their own experts could be seen as a mechanism for building trust among the different parties".

The European Commission Seventh Framework Programme project EAGLE recommended the selection of the following up to date and necessary communication practices for a successful involvement with stakeholders[6]:

(i) Participate in networks with active, empowered citizen communicators;
(ii) Establish 'Science Media Centres' as centralized scientific data services for journalists;
(iii) Translate, simplify and clarify content;
(iv) Use familiar reference points;
(iv) Create and/or support online banks of information that journalists and other stakeholders can consult;
(v) Adapt an institutional communication culture to the actual communication landscape;
(vi) Admit scientific uncertainties and provide balanced information;
(vii) Engage with stakeholders early;
(viii) Know your public's needs and perceptions and how they receive and understand information.

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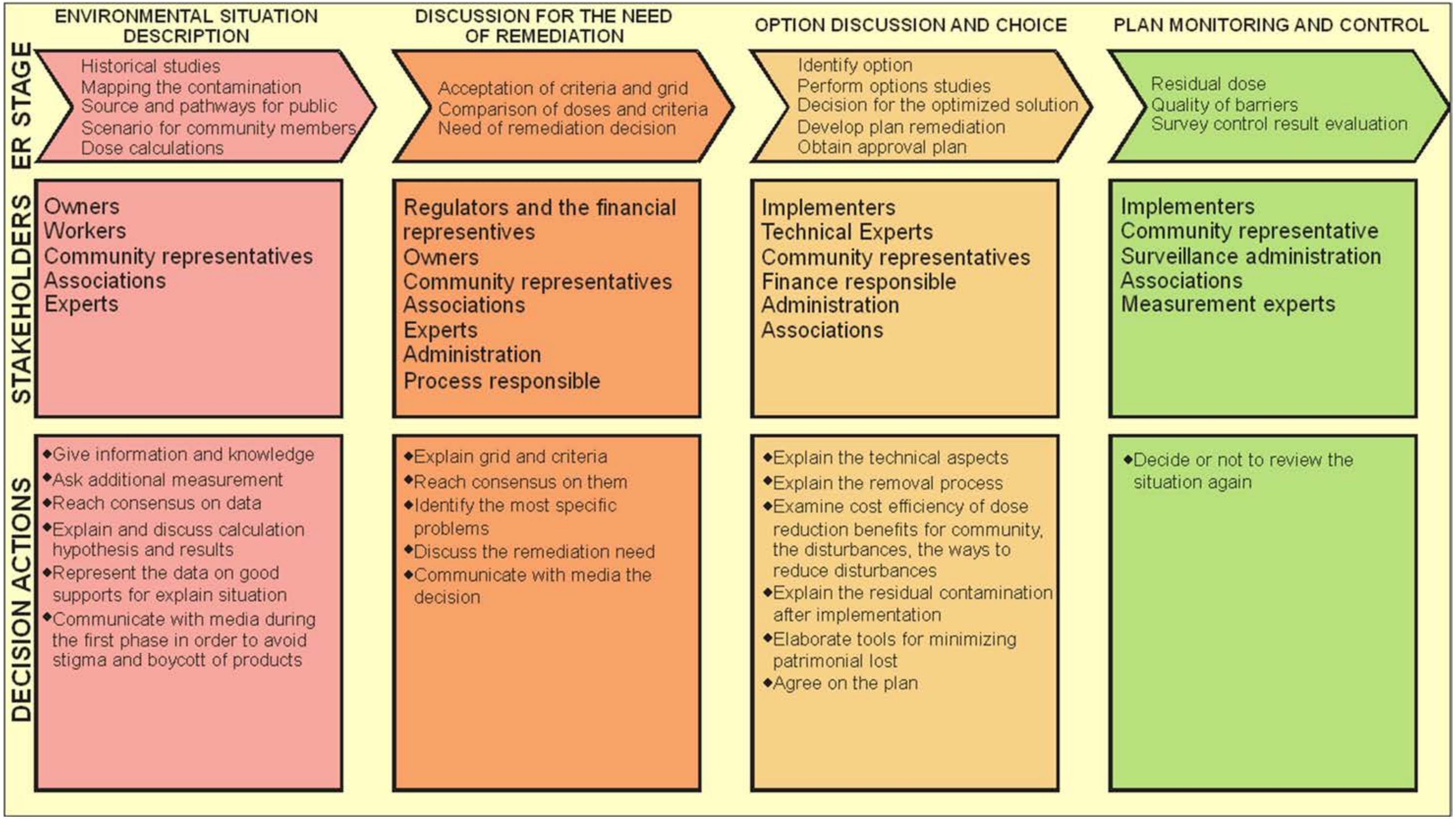
[6] http://eagle.sckcen.be/-/media/Files/EAGLE/EAGLE_recommendations.pdf?la=en&hash=0F6757D218EE7FD448B440F62BA0FED0EF19A8A7.



	ENVIRONMENTAL SITUATION DESCRIPTION	DISCUSSION FOR THE NEED OF REMEDIATION	OPTION DISCUSSION AND CHOICE	PLAN MONITORING AND CONTROL
ER STAGE	Historical studies Mapping the contamination Source and pathways for public Scenario for community members Dose calculations	Acceptation of criteria and grid Comparison of doses and criteria Need of remediation decision	Identify option Perform options studies Decision for the optimized solution Develop plan remediation Obtain approval plan	Residual dose Quality of barriers Survey control result evaluation
STAKEHOLDERS	Owners Workers Community representatives Associations Experts	Regulators and the financial representives Owners Community representatives Associations Experts Administration Process responsible	Implementers Technical Experts Community representatives Finance responsible Administration Associations	Implementers Community representative Surveillance administration Associations Measurement experts
DECISION ACTIONS	◆Give information and knowledge ◆Ask additional measurement ◆Reach consensus on data ◆Explain and discuss calculation hypothesis and results ◆Represent the data on good supports for explain situation ◆Communicate with media during the first phase in order to avoid stigma and boycott of products	◆Explain grid and criteria ◆Reach consensus on them ◆Identify the most specific problems ◆Discuss the remediation need ◆Communicate with media the decision	◆Explain the technical aspects ◆Explain the removal process ◆Examine cost efficiency of dose reduction benefits for community, the disturbances, the ways to reduce disturbances ◆Explain the residual contamination after implementation ◆Elaborate tools for minimizing patrimonial lost ◆Agree on the plan	◆Decide or not to review the situation again

FIG.1. Stakeholder involvement in different stages of the remediation process.

5. CONCLUSION

Governance of risks associated with NORM is challenged by the particularities of radiation (e.g. scientific and societal uncertainties, different perceptions of risks, societal trust issues, awareness issues) and the evolving societal landscape (e.g. social media, active citizenship). The assessment of health and other effects associated with NORM is confronted with the complexity of assessing causal and temporal relationships and uncertainty not only in terms of technical solutions, but even related to system boundaries and ignorance. In other words, better communication, early stakeholder involvement and inclusive decision making related to NORM issues are not only "mission possible" but also "mission unavoidable".

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BUILDING THE BRIDGE FROM BOTH ENDS? COMPREHENSIVE EXTRACTION AND ZERO WASTE STRATEGIES FOR NORM INDUSTRY TAILINGS AND RESIDUES

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Abstract

Led by mining dependent countries such as Australia, Canada and South Africa, the search for a new narrative for the mining and processing sector is currently intense. It is reported that capital expenses write downs in 2015 totalled some $145 billion. For operating expenses, many reports show productivity has declined by 60% in the past 15 years. In simple terms, the old narrative has failed. Whatever else it addresses, the new narrative must encompass an environmentally sustainable, socially acceptable, affordable solution to 'waste'. NORM mining industries such as uranium generate almost 100% 'waste' by volume, either through physical processes (e.g. waste rock) or through chemical processes (e.g. tailings). Phosphate generates a lower mine 'waste' percentage, processing typically 25–35% of the deposit, but during 'wet process' chemical digestion it produces, for each tonne of acid product, five tonnes of the co-product phosphogypsum, which is treated as a residue in some jurisdictions. NORM industry tailings and residues combined run into many billions of tonnes and volume is only part of the problem. The regulatory body — as well as the public — becomes fearful of the NORM legacy content and regulates it as hazardous waste even if that position contradicts the weight of scientific evidence. Bridging to the new narrative starts with a simple but powerful step — stop calling these materials waste. Instead, use value neutral terms such as 'residuals', 'arisings' or (the term used in this paper) 'secondary resources'. Renaming opens the door for value-add uses based on two complementary processing strategies: (i) comprehensive extraction and (ii) ecosystem-wide materials flow management. Comprehensive extraction considers an orebody (or even a whole geological basin) as a single, complex resource and seeks to optimize returns from all the co-located resources, not a single target. Ecosystem materials flow looks at all inputs and outputs and develops a strategy for maximizing the productivity of the system as a whole rather than a single component. As the System of Environmental Economic Accounting (SEEA) and related financial accounting and reporting tools such as Full Cost Accounting are designed to show, when the true costs of the old mining system are captured, the underlying reason for its failure is exposed as being the business model itself.

1. BACKGROUND: FALLING PRODUCTIVITY, SOCIAL RESISTANCE AND WRITE DOWNS

When the concept of comprehensive extraction is applied prospectively to the anticipated 100+ year life-cycle[1] of a major mining and processing project [1] the desired outcome is to maximize the value and quantity of all the mineral resources that can be derived from that project, including tailings and other residues that may subsequently be reprocessed or used as co- or by-products, such that the ratio of resources used to resources rejected is as high as reasonably achievable. When the concept of zero waste [2] is applied retrospectively to that same life cycle, the desired outcome is to minimize the negative social and environmental impacts of mining and processing, and hence the associated negative externality, to the extent that it is as low as reasonably achievable. This paper considers strategies for making sure these objectives are aligned in such a way that the outcome is both environmentally and economically sustainable while also being socially acceptable throughout the four or more generations through which the mining life cycle will pass. Such strategies, if correctly conceived and applied, converge in an ecosystem solution that ensures not only that the mining and processing project itself is logically coherent and consistent, but also places that project in the wider social, economic and environmental ecosystem within which it is situated.

It is a well established business maxim that the best time to innovate or disrupt is at the bottom of the economic cycle. The mining and processing sector is thought currently to be at the bottom of a ‘super cycle’; hence this would be the best time to innovate. Analysis by both Ernst and Young [3] and McKinsey concludes that one indicator of the decline of the sector is the drop in productivity over the past 15 years, a drop estimated overall by both at some 60%. McKinsey summarizes as follows [4]:

> “Over the past decade, mining productivity has declined 3.5 percent per year, meaning that mining companies are 28 percent less efficient in digging and moving a ton of total material today than they were ten years ago.”

The impact of this can be seen most graphically when the performance of the mining sector is compared with the stock market as a whole, where in the period June 2011 to October 2015 mining shrank in value by 60% as compared with a 60% gain in the stock market (see Fig. 1).

In terms of write downs (impairments), SRK puts these at $145 billion since 2011 [5], of which, according to Pricewaterhouse Coopers (PwC), $57 billion were booked in 2014 [6] and according to Bloomberg a further $42 billion were booked in 2015 [7]. In 2016 the write down process continued. In terms of market capitalization, PwC estimates that 2014 saw a drop of $280 billion for the top 40 mining companies [6]. Among the primary causes of such large write downs are {i) the loss of, or failure to gain, the social licence to operate and (ii) the rapidly escalating cost of managing mining projects through the end of life (EOL) phase most notably in regard to decommissioning and waste management. As observed in Ref. [1], it is now unthinkable for a responsible mining project to be initiated at all without a full life cycle cost model being approved including all aspects of EOL management and post-closure remediation.

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[1] A presentation by the CEO of the Roessing Uranium Mine, Namibia to the IAEA Leadership Academy, Walvis Bay, Namibia, 25 November 2015 indicated that the recently agreed licence extension for the Roessing mine extended its expected life to at least 150 years.

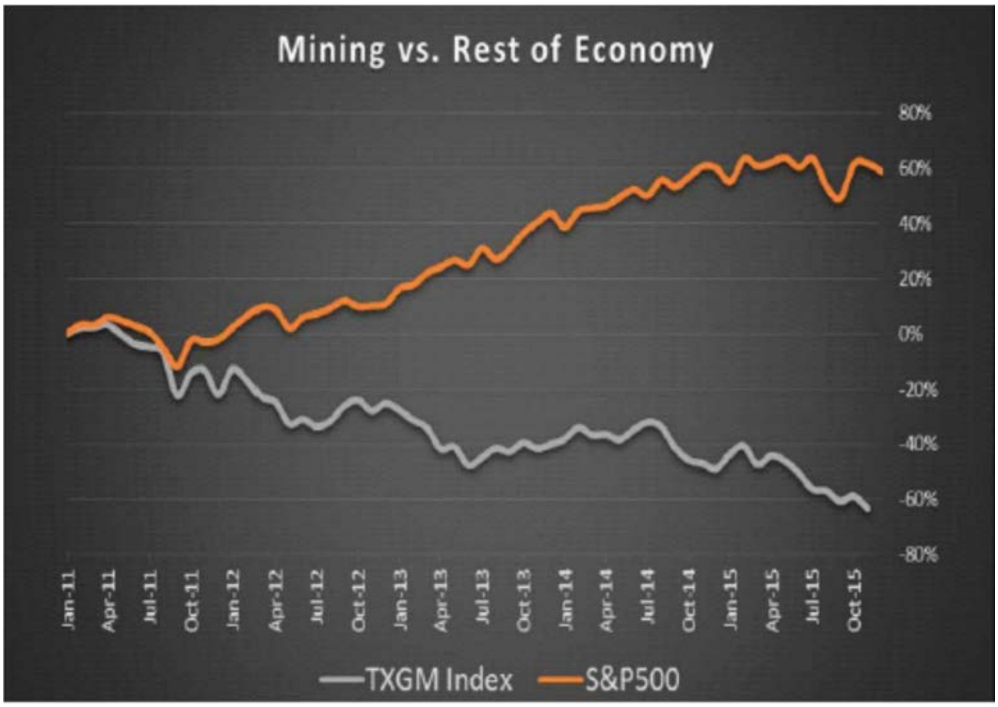

FIG. 1. Mining sector versus S&P Performance, June 2011 to October 2015 [8].

Despite these severe challenges, paradoxically many countries worldwide seeking to diversify their economies (notably away from oil and gas) have focused on significantly expanding their minerals sector. How can these apparently contradictory trends be reconciled? In 2006 the IAEA published a list of industries working with or generating NORM [9]. Our belief is that, of these, two closely linked industries, uranium and phosphates, may give some clues as to the answer, notably: (i) in the way they are starting to look at their potential relationship in terms of co-products and (ii) in the way they both deal with tailings and other residues which traditionally are classified as waste but which are now increasingly being reclassified as secondary resources.

2. A NEW EQUILIBRIUM FOR MINING AND PROCESSING

In a striking intervention ahead of the 2015 South African Mining Indaba, the conference President, Bernard Swanepoel, in his letter of invitation to delegates called for a new mining narrative [10]:

> “Productivity, profitability and positive narrative are all critical and take on a new dimension in 2015 defining the future of the mining sector. This new context is modernisation and it is the bridge to a new mining future. To change the narrative of a 100+ year old industry, in a way that unleashes potential and inspires hope requires that we actually have to change, and that means us!”

What Mr. Swanepoel describes as a new narrative may be redescribed in socioeconomic terms, as defined by Nobel prize winning economist John Nash [11, 12], as a new equilibrium. This equilibrium sees future success in the mining sector as being based on finding a sustainable and equitable balance in mining and processing between environmental, economic and social benefits — these must be shared fairly, transparently and accountably between stockholders (shareholders) and stakeholders. Only when this point of equilibrium is found and adhered to will the productivity decline be reversed and impaired assets return to their proper value.

Referring to the metaphor in the title of this paper "building the bridge from both ends"[2], the two key components are (i) comprehensive extraction and (ii) zero waste. Of these, comprehensive extraction (CX) is a prospective measure, planning, in advance of mining, to maximize returns with respect to both volume and value of the various target resources in an orebody, or more widely in a resource basin. Zero waste (0W), while implemented in parallel with CX, is in effect a retrospective measure designed to ensure that the negative legacy from mining is either zero or as low as reasonably achievable[3]. As Nash showed in his economic theory, these measures combine both forward and backward induction techniques to optimize where the optimum return can be found; and by undertaking this procedure as a 'non-cooperative' rather than 'cooperative' game, the premise is that the equilibrium that results will be particularly resilient because it retains only those outcomes that suit the selfish interests of all parties. This is why the equilibrium can be described as 'win–win'.

3. COMPREHENSIVE EXTRACTION: THE VOLUME–VALUE EQUILIBRIUM AND FOCUS ON CO- AND BY-PRODUCTS

CX has been one of the dominant themes of the work of the Institute of Integrated Mineral Development of the Russian Academy of Sciences (IPKON RAS) since its founding in 1991. The concept was first anglicized by Prof. K.N. Trubetskoy as "comprehensive exploitation and conservation of earth's mineral reserves and resources" [13]. The term was first introduced to the authors of this paper by Dr. Pingru Zhong of the Beijing Research Institute of Chemical Engineering and Metallurgy (BRICEM)[4] at an IAEA technical meeting in Vienna in September 2011, following which Prof. Trubetskoy's vision of an equilibrium between resource use and conservation was adopted under the rubric 'comprehensive extraction'. CX seeks prospectively to maximize:

- The operational mining objective of volume recovery (including productivity, recovery efficiency and hence resource conservation);
- The commercial mining objective of deriving social and economic value of all the target minerals in an orebody (or wider energetic resources of a whole basin [14]).

For both objectives, optimizing current technologies for mining and introducing innovative or disruptive ones to enhance resource conservation, with an associated emphasis on building competence based capability in the use of such technologies and techniques according to Trubetskoy, remain overarching imperatives [15].

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[2] Euclid's isosceles triangle theorem is also known as the 'pons asinorum', the bridge of donkeys, because Euclid (Elements, Book 1, Proposition 5) appears to have believed that understanding this theorem was the bridge to the remainder of his work. We argue that constructing such a new equilibrium will be the bridge to the new, sustainable mining narrative.

[3] The legacy is defined as negative because the proposition of indefinite stacking of wastes is logically absurd and to sterilize land indefinitely is in effect infinitely expensive. It is clear that future generations will at some time want to remedy what has been left behind, the only open question being when this occurs.

[4] See http://www.bricem.com.cn/en/?optionid=409.

A mine does not close because it is running out of mineral resources. It ceases operation after exhausting the resources above a certain cut-off grade. When the project is required to process lower grade resources, the metal content of the produced ore decreases, sometimes drastically, making the overall economics negative. Production can be increased only by mining and processing more ore, which requires additional mining, transport and processing capacity. However, metal production could still remain lower in spite of large capital and operational expenditures. CX initially looked into three aspects: (i) the technical feasibility of how the efficiencies and recoveries can be increased, or how mining and processing losses could be minimized, (ii) how lower grade ore could be mined and processed with increased efficiencies; and (iii) how mining productivity could be increased by recovering multiple metals or materials. This approach requires re-evaluation of mining projects along multiple lines:

- Geological knowledge and methods of assessment;
- Technology options to recover multiple materials and their value addition;
- Socioeconomic parameters that can be made favourable.

These challenges are difficult to address using a range of different mineral management and reporting systems whose origins long predate the demands of sustainable development and whose scope is predominantly to protect the interests of private investors, but such systems remain in common use today. Recognizing this problem, the United Nations Framework Classification (UNFC–2009) has been designed to bring consistency and comparability to resource management while also for the first time enabling meaningful comparisons to be made for resources of all types, including minerals, oil, gas and renewables [16].

Other initiatives to promote sustainability in mining are gaining prominence. Some companies are using data mining or big data analytics to improve efficiency. Others, such as Cameco, are deploying autonomous vehicles, such as for mining uranium underground at Cigar Lake [17], or are developing new techniques for 3D mine face scanning [18], portable field based[5], in-line analysis of mine samples[6] either to make mining itself more efficient or to better manage the orebody during beneficiation. There is a complete ‘digital reinvention’ to improve productivity, reducing costs to operate, and to prevent potential mishaps by closely tracking everything from water quality to necessary equipment maintenance.

As the crisis of confidence in the mining industry, first diagnosed by PwC in 2013 [19], has deepened, a natural convergence has emerged between the principles of CX, the rapidly changing socioenvironmental context within which mining operates and the long term objectives of sustainable development. As has been well articulated by SRK, this has had the consequence of refocusing the finances of the industry on the role of by-products and co-products in the definition and calculation of its net present value [20], and this process is now driving the adoption of CX into industry good practice, for both new and existing long term projects.

4. ZERO WASTE

A zero waste initiative has been embarked upon in Canada by the Canada Mining Innovation Council (CMIC) to bring a staged, concerted approach to the desired zero waste solution [2]. The problem statement is as follows:

> “Base metal mines typically recover less than 1% of the volume of rock extracted. Most gold deposits recover less than 0.001%. Typical cut off grades for uranium mean that 1 million tonnes of rock mined yields about 500–1000 tonnes of uranium ore. The result

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[5] See for example TerraSpec, http://www.asdi.com/products-and-services/terraspec.

[6] See for example TOMRA, https://www.tomra.com/en/solutions-and-products/sorting-solutions/mining/why-tomra/.

> is the extraction of huge volumes of rock that end up either in mine waste piles or tailings ponds. Both represent a major part of production cost and of the mine footprint that must be managed for their potential environmental impact after mine closure.
>
> “The Canadian mining industry’s greatest challenge then is how to more efficiently extract the desired commodity through the minimal displacement of host rock, and more effectively managing mine tailings that continue to be produced” [2].

The CMIC clearly understands that a change as profound as zero waste can only be achieved by mobilizing all stakeholders — industry, academia and government — to develop innovative, potentially disruptive and progressively more efficient extraction methods:

> “The CMIC technical groups are addressing a number of cross-disciplinary and linked initiatives leading towards reducing the mining footprint. The groups have identified targets and are developing innovation priorities that will lead to significant reductions in mining waste in the next 5 years and move towards net zero waste in mining and mineral processing in 10–20 years.”

The approach applies a full life cycle model to the mining project into which intermediate milestones are inserted at which a thorough performance review is required. Performance indicators are dynamic in nature and track the capital value process through: (i) more efficient definition of new ore discoveries, (ii) the demonstrable application of more effective in situ mining methods, for example to minimize waste rock production, (iii) the application of closed system processing to reduce wasteage of water and energy, and (iv) the reprocessing of mine tailings, or their use as a co-poduct, in line with the waste hierarchy principles [21] towards a benign, saleable product. CMIC concludes:

> “These inter-disciplinary and inter-sectoral consortia will minimize overlap and focus joint efforts on the mutually understood end point of zero mine waste” [2].

Zero mine waste is achievable through innovation, the adoption of new technologies and mostly through stakeholder involvement for a collective pursuit of efficient use of primary and secondary resources to benefit both the mining industry and its indirect and direct beneficiaries.

Australia also recognizes that mining activities throughout their life cycles are prone to generate very high volumes of waste. Therefore, there is a requirement for modernization steps to eliminate this waste which are very similar to those envisaged by CMIC. The case for a comparable approach for Australia has been set out by W. John Rankin [22]:

> “Company behaviour has moved in recent decades from complying with regulations to corporate social responsibility. In the next decades, it will need to move progressively to ‘closing the loop’ strategies to dramatically reduce the quantities of wastes. The drivers for change have moved from being almost exclusively profit to include regulations, stakeholders and increasingly to changing social values. In parallel, the materials cycle focus has shifted from a narrow focus on products towards including co-products. Increasingly, focus will shift to the entire materials cycle and, ultimately, to the entire economy”.

The emphasis on co-product use is pivotal for the future and underlines why such an approach is recommended for energy related resources such as uranium in particular.

5. WASTE HIERARCHY

The EU Waste Framework Directive [21] establishes a strategy for prioritizing management of 'waste' in a hierarchical form as follows:

(i) Prevention,
(ii) Reuse,
(iii) Recycling,
(iv) Processing or recovery,
(v) Disposal.

Of these, the least favoured option is disposal. It is self-apparent how well both CX and 0W align with this directive and the hierarchical decision gates for waste management it mandates. The operational resource management consequences of applying the waste hierarchy (see Fig. 2) are briefly explored below.

FIG. 2. The EU waste hierarchy.

6. NEW ECONOMIC RESOURCES

If the waste hierarchy can be characterized as a 'push' factor for change, driven by the regulator and by policy, the development of the concept of 'new economic resources' and its application notably to the secondary resources generated by mining processing, tailings and other residues, is a balancing 'pull'. Accordingly, the International Accounting Standards Board (IASB), in a new accounting term defines 'economic resource' as: "a right, or other source of value that is capable of producing economic benefits" [23]. Such rights of course include any intellectual property rights developed by the entity managing and using secondary resources in innovative ways, but equally rely on the concept of competency in the execution of those rights to achieve the value for them that is stated in the accounts.

7. ENVIRONMENTAL–ECONOMIC ACCOUNTING

The concepts of 'new economic resources' and 'environmental–economic accounting' are reciprocal, and in effect a further expression of the Nash equilibrium. Both ideas have their roots in the Brundtland Report: Our Common Future [24], which launched the whole sustainable development movement, a provenance set out clearly in the introduction to the System of Environmental–Economic Accounting (SEEA) [25]. SEEA consists currently of two primary documents (i) the Central Framework [25] and (ii) the Experimental Ecosystem. While the initial emphasis of the SEEA Ecosystem component was on the biological definition of the ecosystem, based on the service biology concept whereby ecosystems must be made sustainable to serve the needs of current generations without comprising the ability of future generations to meet their needs, now it is recognized that the understanding of ecosystem must be widened to include industrial ecosystems as a whole, beginning of course with naturally occurring resources whether animal, vegetable or mineral. The glossary of key terms in the SEEA Central Framework document includes many such terms, making this process relatively straightforward.

Why are the use and recycling of secondary resources from NORM industries, such as phosphates and uranium, the 'locus classicus' for new economic resources? The SEEA provides a way of explaining this in terms of value derived from what the SEEA terms the 'release of residuals' [25].

8. RESIDUALS

The SEEA defines residuals as follows:

> "Residuals are flows of solid, liquid and gaseous materials, and energy that are discarded, discharged or emitted by establishments and households through processes of production, consumption or accumulation."

Such a definition reinforces the belief of the authors of this paper that CX must be applied to whole geological basins not just for the mineral content but also for oil, gas and related liquid resources, which are themselves also classed by the IAEA as NORM industries [9]. In the introduction to the SEEA [25] the authors comment on the evolution of the meaning of the term 'residual' and its impact on accounting systems as follows:

> "There is now a clearer delineation of the boundary of physical flows with respect to the production boundary of the System of National Account (SNA). In particular, (a) all cultivated biological resources are considered to be within the production boundary, (b) all flows to controlled landfill sites are treated as flows within the economy, and (c) there is a consistent treatment of so-called natural resource residuals (referred to in the SEEA-2003 as "hidden" or "indirect" flows). In addition, flows relating to the inputs of energy from renewable sources are explicitly recognized in the SEEA Central Framework, and a definition of solid waste has been provided".

For the SEEA, the concept of 'residual' is integral to life cycle resource management, which is defined as follows:

> "Resource management activities are those activities whose primary purpose is preserving and maintaining the stock of natural resources and hence safeguarding against depletion."

Such a definition is clearly consistent with the Trubetskoy principles of CX and those of 0W. Once applied it invokes a logical next step in the accountancy process, that of revaluing what previously had been termed waste — and hence classed as liabilities — into secondary resources which are now classed as assets. Revaluation is hence defined by the SEEA as follows:

> "Revaluations relate to changes in the value of assets due to price changes and reflect nominal holding gains and losses on environmental assets. The nominal holding gain for environmental assets is calculated in the same way as for non-financial assets, as the increase in value accruing to the owner of the asset as a result of a change in its price over a period of time."

It is clear that taking a positive and proactive approach to managing value release from residuals has a significantly beneficial impact on the social licence to operate, especially in NORM industries where the perceived risks from radioactive materials compound negative public attitudes to mining and processing in general [26].

9. THE PHOSPHATE RESOURCE LIFE CYCLE: PHOSPHOGYPSUM AND URANIUM

The phosphate industry illustrates very well why CX, 0W and SEEA must take a central place in the new narrative of mining and processing because they are the fundamental tools available to the industry to manage the new environmental–economic equilibrium on which its future rests. These principles can briefly be applied to (i) the recovery of uranium and rare earth elements from the ore during wet process phosphoric acid production, and (ii) the sustainable management and use of phosphogypsum (PG) as the primary residue of the phosphate industry.

With regard to the extraction of uranium and rare earths from phosphoric acid (UxP), the IAEA has been very active since 2009 in supporting efforts to regenerate and enhance this mature and commercially proven technology within the CX framework. A summary of the current state of UxP is provided in IAEA Safety Reports Series No. 78, Radiation Protection and Management of NORM Residues in the Phosphate Industry [27] and a very detailed IAEA Manual on this topic is currently in the publication process.

With regard to PG, this of all NORM industry residues is perhaps the greatest in quantity with an annual production of 215 million t and stockpiled holdings of some 4 billion t worldwide. The change of fortunes of PG in the global phosphate industry since Safety Reports Series No. 78 was published in 2013 [27] is perhaps the most striking illustration in recent times of all the principles in this paper converging. It effectively hangs on one word 'co-product'. Since the promulgation in 1989 by the US Environmental Protection Agency (EPA) of the "Phosphogypsum Rule" [28], PG had been classed as a hazardous waste on radiological grounds and was subject to a mandatory stacking requirement amounting to permanent disposal. The then dominance of the United States of America in the phosphate industry meant that this rule had an impact well beyond that country. IAEA Safety Reports Series No. 78 [27] refers to PG as a co-product in the following way:

> "A particular example in this regard is phosphogypsum, a co-product of phosphoric acid production that, because of the very large amounts involved, is stored in aboveground engineered containments known as 'stacks', often for indefinite periods […]. Evidence shows that, with [appropriate] controls in place, there is no necessity for additional regulation for purely radiological purposes. Future liabilities associated with the continued presence of large phosphogypsum stacks place a considerable burden on future generations. This, together with the increasing rate of phosphogypsum production, provides a very compelling reason for creating a regulatory environment that is conducive

> to identifying and promoting further ways for safely using phosphogypsum as a product rather than having to manage it as a waste.”

Following the publication of this IAEA Report, the International Fertilizer Industry Association (IFA) commissioned a report entitled Phosphogypsum: Sustainable Management and Use, which was published for member companies in March 2016 [29]. It is clear from the findings of the IFA report that by removing the radiological objection to the use of PG as a result of it not being supported by the evidence, not only was the industry and the regulator encouraged to “identify and promote ways of safely using PG as a product”, but the social and market acceptance of PG as a secondary resource caused exactly the value release proposed by SEEA with the consequent benefit to the industry of placing vast quantities of PG in the asset column rather than the liability column on the balance sheet. In the case of the 4 billion t of PG currently stacked, its categorization as a secondary resource rather than waste could generate, in terms of direct costs and land values, an aggregated value swing of the order of $70/t. This equates to a capital value change (revaluation) of up to $280 billion, a figure that bears directly on the asset value of the global phosphate industry. How such value release can in practice be achieved is now being actively pursued worldwide, with the use of PG in 2015 already representing some 25% of production [29].

The process of integrating such an approach into the SEEA ecosystem is also straightforward. The SEEA defines ecosystem accounting as follows:

> “Ecosystem accounting is a coherent and integrated approach to the assessment of the environment through the measurement of ecosystems, and measurement of the flows of services from ecosystems into economic and other human activity.
>
> “The use of an accounting framework enables the stock of ecosystems — ecosystem assets — and flows from ecosystems — ecosystem services — to be defined in relation to each other and also in relation to a range of other environmental, economic and social information.
>
> “Ecosystem accounting aims to shed light on the non-market activity that relates to ecosystems and integrate this information with relevant market related data.” [7]

As this paper has attempted to show, the bridge that can be built by applying CX and 0W principles to secondary resources enables SEEA ecosystem accounting not only to ‘shed light’ on non-market activity, but to converge the SLO with the Legal Licence to Operate, in exactly the way advocated by SRK in regard to how to reconstruct the financial instruments for the mining and processing sector [8]. The outcome, as UxP and PG illustrate, can be a very significant release of value from residuals resulting in triple bottom line (social, environmental and economic) gains.

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[7] An example of significant non-market activity over which the operator has no control is the impact of the large sinkhole which opened up on 27 August 2016 at the Mosaic Mulberry Stack, putting the industry’s social licence to operate at risk as a result of local protests, see http://wfla.com/2016/09/17/contaminated-well-concerns-prompt-protests-after-mosaic-sinkhole-incident/.

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CASE STUDY IN THE MUNICIPALITY OF IPERÓ REGARDING THE BRAZILIAN MULTIPURPOSE NUCLEAR REACTOR IMPLEMENTATION AND PUBLIC ACCEPTANCE: WOULD IT BE A MATTER OF COMMUNICATION?

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Abstract

The city of Iperó will receive the implementation of the first Brazilian multipurpose nuclear reactor (RMB) for research purposes and producing radioisotopes for health and industry, with a strong social appeal. On the other hand, it is known that public opinion about nuclear technology has changed over its history, with the emphasis shifting from the scientific perspective (relatively favourable opinions) to the political perspective (less favourable opinions). In 2013, there were public hearings in the cities of São Paulo, Iperó and Sorocaba to inform the population about the RMB project, which was disseminated through several media resources. From the results of public hearings, as well as related news in the local print media, it would appear that the general population does not regard the implementation of the RMB as being acceptable or favourable. Considering this perspective, during the period April 2015 to January 2016, part of the Iperó city population was interviewed through a semi-structured questionnaire that aimed to investigate the social representations and perception of risks related to the implementation of the RMB. The results demonstrated that interviewees suffered from a lack of information about the project, thereby directly influencing the implementation and acceptance of the RMB since the benefits were not easily perceived.

1. INTRODUCTION

Radioactivity is a natural phenomenon that has persisted throughout the evolution of the planet. However, knowledge of its existence came about relatively recently through the accidental discovery of Becquerel in 1896 when, in his experiments using uranium salts, he noted that the salt darkened photographic film. Afterwards, Marie Curie and her husband, while investigating the properties of uranium, discovered other radioactive elements such as polonium and radium, which led Marie to the hypothesis that the emission of rays by uranium compounds could be an atomic property of this element. This important fact would trigger not only a revolution in our society but also the most controversial 'love–hate' relationship that has built up throughout the history of contemporary science. During this period, people experienced the 'belle époque' in Europe, where science occupied a prominent place, with new inventions and discoveries. The great interest aroused by radioactivity, as discussed in Ref. [1], led to the appearance of numerous theories aimed at justifying the application of therapies and supply of various products with added radioactivity, from radioactive drinks to therapeutic treatments involving radiation.

However, 'social representation'[1] related to radioactivity changed from positive to negative after the Second World War, when the atomic bombs 'Little Boy' and 'Fat Man' were detonated over Hiroshima and Nagasaki, respectively, in 1945. The change to a negative social representation was further accelerated by the accident at the Chernobyl plant in 1986. On the other hand, there is an understanding that radioactivity is present in our daily lives more than we initially imagined, whether it be associated with naturally or radioactive materials used in

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[1] Social representation as an established theory emerged in 1969 through the work of Serge Moscovici. It is an alternative approach to understanding the everyday behaviour of individuals and their social groups. The approach was able to contribute significant knowledge about public perceptions on different topics.

energy production, medicine in research, or among other relevant applications. We acknowledge that nuclear technology also brings numerous benefits.

Within this perspective, the proposed installation of the Brazilian multipurpose nuclear reactor (RMB) in the municipality of Iperó can be regarded as advantageous in view of the numerous benefits that this investment will bring to society. In this sense, since the project was announced in public hearings, why were so many residents of Iperó against the project? How to solve this issue? Where to start? In fact, this is not exclusively a Brazilian issue — there are many efforts in various countries to address risk perception and the social representation of the general population on the nuclear issue [2–5]. In the nuclear area, according to Ref. [6], the study of risk perception is crucial and several factors are used to explain it.

It is pointed out in Ref. [7] that risk perception has appeared strongly in politics since the 1960s in the sense of opposition to technology, especially nuclear technology, even when recognizing that risks in everyday life (such as smoking or drinking and driving) are greater than those associated with a nuclear power plant. Therefore, when there is a rejection of a type of technology and acceptance of others, even though all of them may have their risks, this is when the risk perception study plays a fundamental role. Reference [8] demonstrates the existence in risk perception of an influence-denominated 'cultural risk theory' [9][2], which considers that the risks do not exist outside of the subject to be quantified. But because the risks can be subjective and influenced by psychological, social, institutional and cultural factors, the psychometric paradigm[3] assumes the possibility of quantification of these factors as research tools, by means of a suitable design of risk perception. In addition, in studies aimed at investigating the risk eerception of a particular group or population, it is common to use different evaluation tools, which aim to measure the reality of an object under study. However, to ensure those results are reliable and close to reality, it is necessary to develop or use appropriate data collection instruments [10].

In this study, which is aimed at elaborating the risk perception of the population of Iperó regarding the implementation of the RMB, we used the Likert scale. We adopted the scale from 1 to 7, in most of the questionnaire, which aims to verify the agreement and disagreement on the subject matter, as discussed in Ref. [11]. Within this perspective, this paper discusses the questions about the perceived benefits with the arrival of the RMB, the aversion to the project and the level of respondents' confidence in various institutions. Getting to know the public opinion of local residents related to the arrival of the RMB to the municipality can help with the acceptance of the technology, improve its application and assist in recognizing the importance of social participation in decision making processes, thus helping to improve the future prospects for the nuclear area.

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[2] Cultural risk theory addresses the role of culture in the development and acceptance of risks produced by modern society [9].

[3] In short, the psychometric paradigm encompasses a theoretical framework that assumes risk to be subjectively defined by individuals who may be influenced by a wide array of psychological, social, institutional, and cultural factors. The psychometric paradigm assumes that with appropriate design of survey instruments many of these factors can be quantified [8].

2. METHODOLOGY

2.1. Choice of city

The RMB construction project was established as a goal of the Action Plan on Science and Technology and Innovation of the Ministry of Science, Technology and Innovation in 2007. The implementation of the reactor will be in the municipality of Iperó in the State of São Paulo, occupying an area of approximately 2 million m^2 and adjacent to the Central Experimental ARAMAR (CEA), popularly called Aramar (Fig. 1). Elevated to a municipality by State Law No. 8092 on 28 February 1964, the city of Iperó is the name of one of the rivers bordering the territory of the municipality and means, according to the official version, “deep water and riots.” It is located 25 km from Sorocaba and 116 km from São Paulo, the State capital. According to the SEADE [12] and the IBGE [13] in 2016 and 2015, respectively, Iperó covers an area 170 km^2 with a population density[4] of 185 inhabitants per square kilometre. The city’s population in 2015 was 31 531. It has 706 registered and active companies, with an average income of the population of about 3.2 times the minimum wage in 2010 with an HDI of 0.719. It also has 14 basic education schools, 11 municipal networks and two of the state IBGEs (2015) [13].

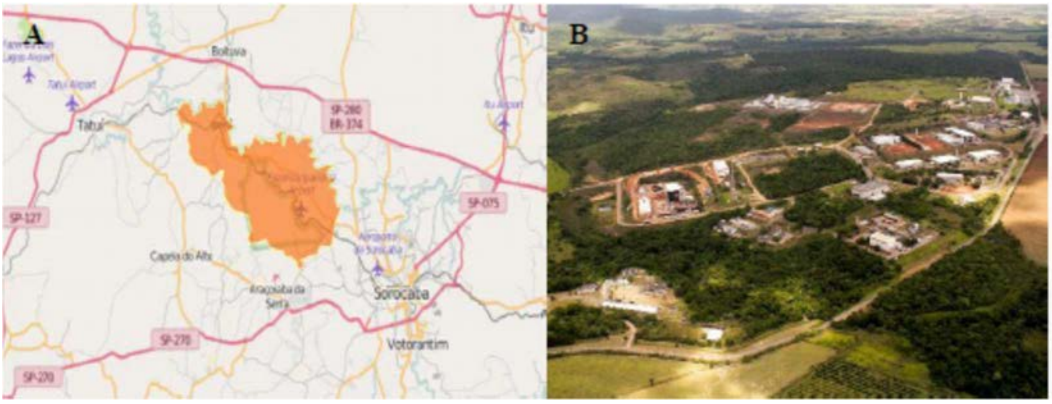

FIG. 1. Map of the city of Iperó (left); local implementation of the RMB (right).[5]

2.2. Data collection

This study was exploratory in nature, in which the answers found to the initial assumptions provided a preliminary knowledge of the subject, little explored in the region, in order to support new issues to be investigated in the unfolding of this work. So it is qualitative in the form of a case study, as recommended in Ref. [14].

The development of the questionnaire used in the interviews was based on the work described in Refs [15–17], with the objective being to learn about the attitudes, beliefs, level of risk denial, and trust in institutions and in the accumulated scientific knowledge. Moreover, belief in the possible community stigma due to implementation of the RMB, environmental awareness and the possible sources and share of social representation were also objectives, by using the Likert scale, as mentioned above.

This paper presents an extract from a dissertation on the results obtained in the interviews of Iperó municipality, related to the involvement of the participants with the arrival of the RMB to the city, analysing the level of participation in public hearings and the feeling of safety (or lack of security) with the implementation of the reactor. Also presented is an evaluation of participants regarding the possibility of risk to persons and to the municipality, the individual

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[4] Number of residents living in a geographical unit at any given time in relation to the area of that unit.

[5] Source image : https://www1.mar.mil.br/amazul/?q=projetos.

judgment of participants related to self-protection in the case of an accident in the RMB facilities, as well as the risk perception related to the city's image and the safety of residents.

The questionnaire to the local population took place in Iperó between the second half of 2015 and the first half of 2016, during which period the participants were interviewed in the local market, health clinics, street or homes. The interviews were made over a range of ages of individuals, both residents and municipal workers. We conducted 198 interviews in the city, in which the average age of the respondents was 37, with 55% being females and 45% males. Some 76% of the respondents lived principally in the urban area. Among the residents who answered the survey, 76% were economically active and only 24% were economically inactive (including retired individuals). Analysing the level of education, it was found that most respondents had higher education (going to college), complete or incomplete, or had high school education, each of which accounted for 39% of respondents. Participants with basic education accounted for 15% of respondents, while 6% reported having graduate and 1% did not answer the question.

3. RESULTS AND DISCUSSION

Law 6.938/81 provides for the National Environmental Policy and CONAMA Resolution No. 001/86 governs the process of environmental licensing [18][6]. Public hearings are utilized to present the results of environmental studies and create opportunities for people to ask questions about a particular venture. In the municipality of Iperó, a public hearing was held for presenting and debating the implementation of the RMB [19]. In 2013, there were three public hearings, one in Iperó, one in Sorocaba and another in São Paulo. The hearing in Iperó attracted little involvement of the population, even though the organizers had promoted it by disseminating information throughout the city.

This result for public hearings was also reflected in the responses obtained during the questionnaires, as many residents claimed not to know about the RMB implementation project, were uninformed about the public hearings and, especially, were unaware of the purpose of the reactor. Responses to the question in the questionnaire that sought to know the involvement of respondents with the arrival of the RMB to the municipality showed little involvement with the project, as well as a lack of information and low interest in participating in the public hearings occurring during the period (Fig. 2). On first being approached with the questionnaire, many participants said they did not know about the project and did not receive information about the event, even though the report submitted by the Comissão Nacional de Energia Nuclear (CNEN) contained photos of dissemination advertisements and news in the local media [19].

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[6] A preventive legal instrument that should be observed prior to installation of any enterprise or potentially polluting or environmentally degrading activity [18].

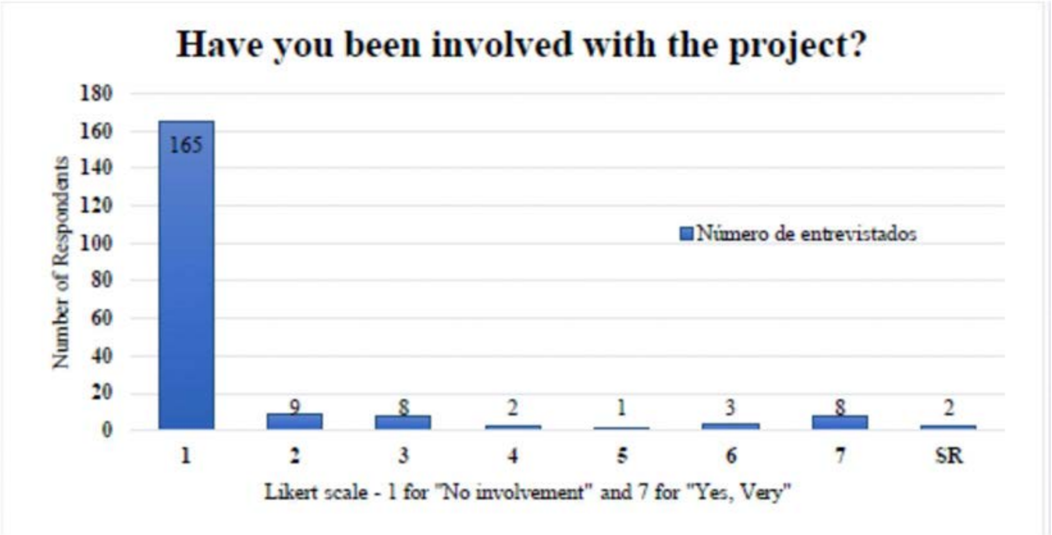

FIG. 2. Judgments of the respondents about their own involvement in the implementation of the RMB in the city.

Responses to the question that was aimed at establishing the feelings of the respondents about the arrival of the reactor to the city showed that in most cases the participants did not feel safe with the implementation of the RMB in Iperó (Fig. 3). This concern was also expressed in the public hearing held in Iperó, as can be seen in the document presented by the CNEN [19], indicating that there was still a gap between the public and experts, even after almost two years of completion of the public hearings. It indicates that the public hearings, as well as other possible interventions, were not successful in changing the risk perception of a group from the studied population. As noted earlier, the report on the public hearings highlighted general concern regarding the safety of RMB deployment in the city, as illustrated by the response of a participant in the public hearing held in Iperó, indicated on page 95 of the report [19]:

> "Are we going to have security if something goes wrong with the RMB? Will we be warned in time to evacuate our homes, or will we be left to ourselves? It is a nuclear reactor, is it not? For better or for worse, Bacaetuva is a neighbourhood next to the RMB project with a population 358 inhabitants".

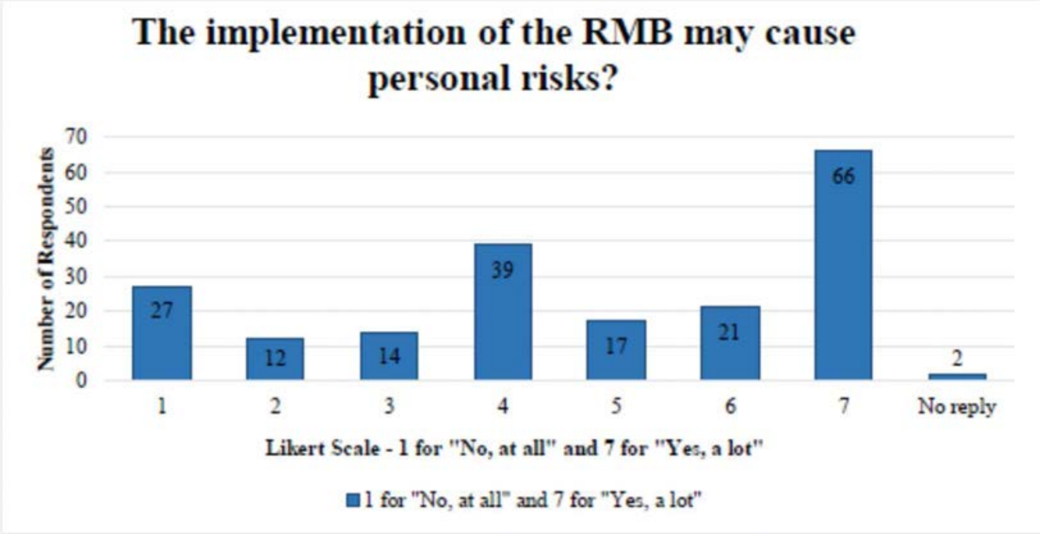

FIG. 3. Feelings related to personal safety related to the implementation of the RMB.

To check on the perception of personal danger from the reactor, respondents were asked to judge the possibility of personal risks related to the implementation of the RMB. The results are shown in Fig. 4. According to the data obtained, we can say that the risk perception of respondents is high due to the higher number of answers scoring 4 or more on the Likert scale. On page 100 of the report on the public hearings [19], a participant raises the following question:

> “...I understand that it is safe, but the others are lay people — what do you have to say in order to explain things in non-technical terms so that people can stay calm?”

The experts assured the local resident and the others present in the public hearing that environmental education programmes would be implemented in order to work on the risk perception didactically, so as to overcome possible risks of misunderstanding. Up to the first half of 2016, no environmental education programmes were embarked upon in the city and the interviewed public did not know about the project.

FIG. 4. Evaluation of the respondents about the possibility of personal risks.

Another issue addressed with members of the local community was their personal assessment regarding the possibility of a risk to the municipality of Iperó with the arrival of the RMB. The answers to the question are presented in Fig. 5. The answers provided to this question show a high risk perception because the majority of respondents preferred to answer the question with a score of 4 or more, mainly choosing the end of the scale (a score of 7) for the possibility of some risk to the municipality. Again, the data may show that the gap in communication between the public and the experts did not decrease after the public hearings, highlighting an urgent need to clarify the project for the population.

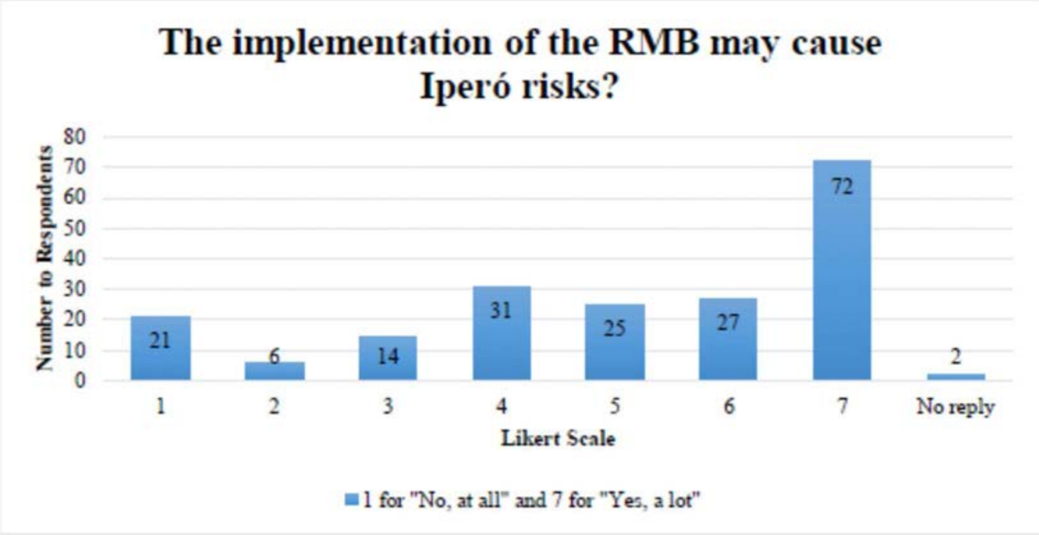

FIG. 5. Evaluation of respondents as to the possibility of negative events occurring in Iperó using the Likert scale.

Initially, the results showed a negative risk perception of the population regarding the arrival of the RMB in Iperó. However, some issues included in the questionnaire were aimed at finding out about the respondents' view of the perceived benefits of the reactor to the country. Again, the respondents were asked to respond using the Likert scale. The results are shown in Fig. 6. Although 20% of respondents thought that there would be no benefit to Brazil, the vast majority of respondents (66%) had a low risk perception regarding the benefits that the RMB could bring to the region, choosing scores of 4 or more on the Likert scale. At the same time, on analysing separately the percentages associated with each score on the scale, the highest percentage was associated with a score of 7 (high benefit to the country).

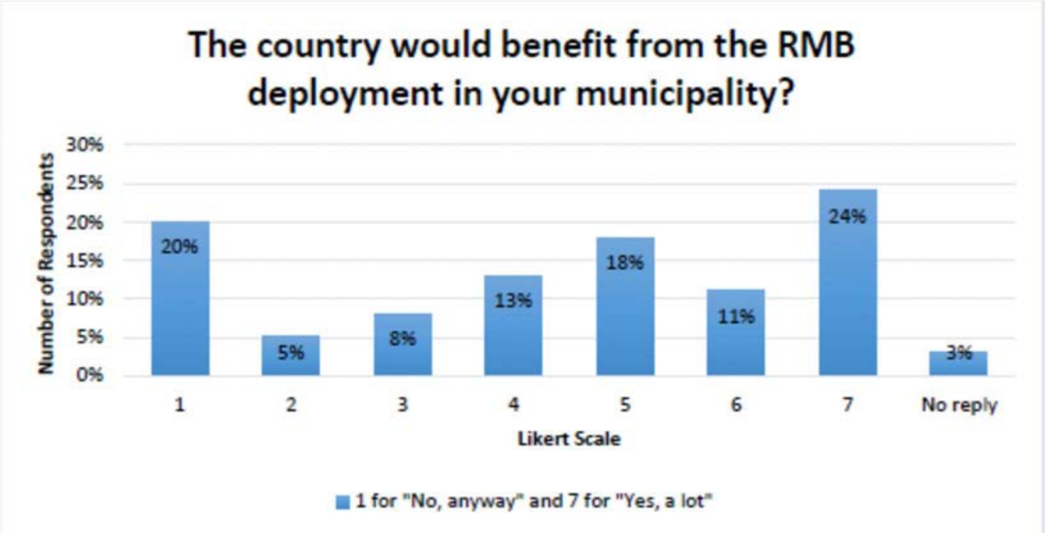

FIG. 6. Evaluation of respondents regarding benefits to the country with the arrival of the RMB at Iperó.

Respondents were asked if Iperó trade would be adversely affected with the arrival of the RMB. The evaluation again was based on the Likert scale. The results are shown in Fig. 7. In this evaluation we can say that the respondents did not associate the arrival of the RMB with possible changes in trade, with most answers to the question being a score of 4 or less on the Likert scale (81% of respondents). Around 43% of respondents gave a score of 1, which could facilitate the acceptance of the reactor presence in the city if the issue were to be addressed in a more efficient way for locals.

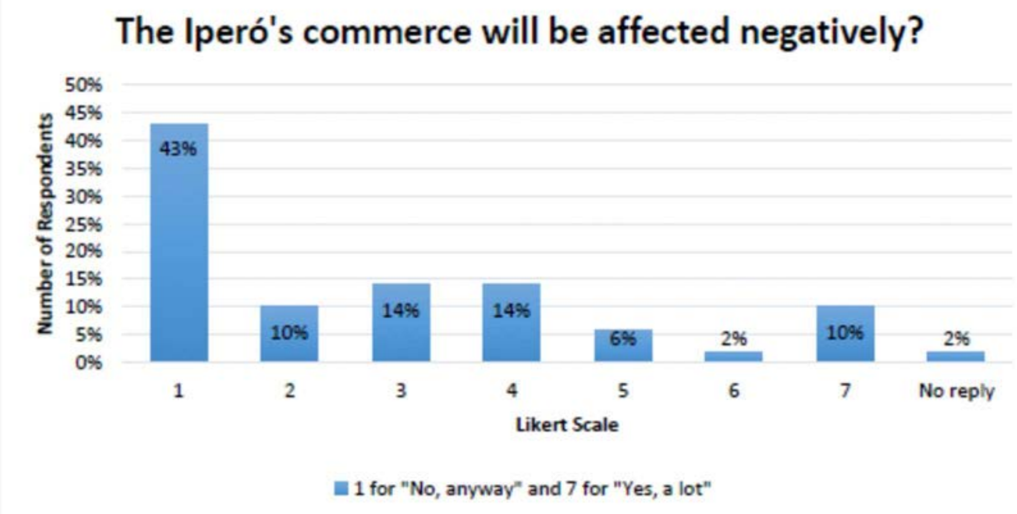

FIG. 7. Assessment of respondents regarding the negative impact on the city's trade.

Another question in the interviews referred to the feeling of pride in knowing that the municipality of Iperó would receive the RMB and if the future would be a credit to the place. The results are shown in Fig. 8. In this evaluation, the highest percentage (32%) was for a score of 1 on the Likert scale, suggesting strongly that the reactor would not induce a feeling of pride within the city for most respondents. If we analyse the set of answers, we can also say that most of the respondents judged that the reactor would not be a source of pride because they opted for scores of 3 or less.

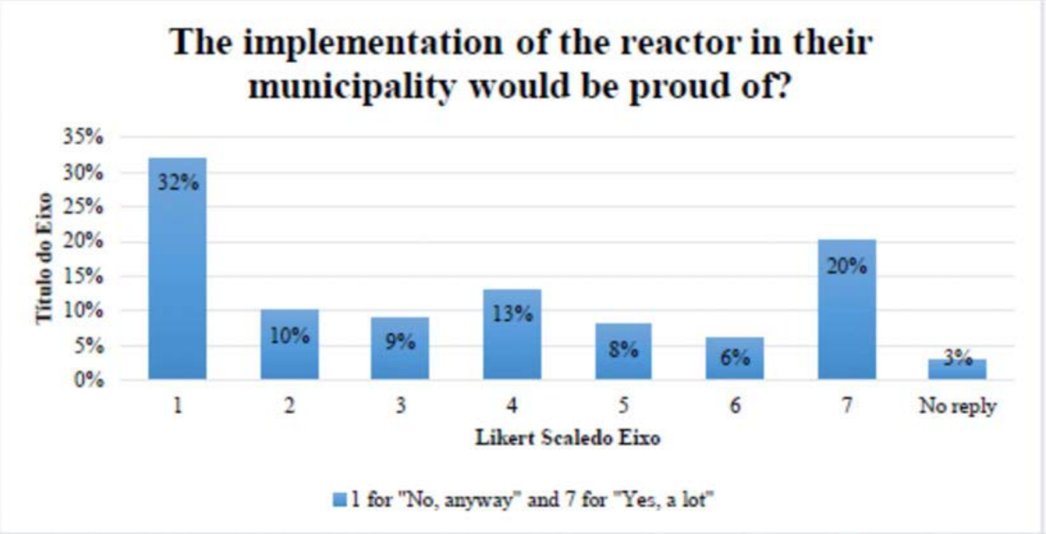

FIG. 8. Assessment of respondents on a feeling of pride regarding implementation of the reactor.

From the responses to this questionnaire, it can be concluded that there would be some acceptance of the deployment of the RMB in the city because of the benefits to the country. On the other hand, the perception among respondents was that such deployment would not bring economic benefits to the local community and would not be a source of pride among the population. These negative views could easily be changed if there were to be greater knowledge among, and easier access to, local residents, as well as more frequent interventions in order to allay their concerns.

4. FINAL CONSIDERATIONS

The risk perception related to nuclear technology changed after the Second World War, when such technology started to be seen in a negative way. Since then, the technology has also come to be used for energy production in several countries. According to some experts, nuclear power causes less impact on climate change and may therefore be an option for sustainable energy systems [20], at least while making the transition to other energy sources. Over the years, however, there have been failures in nuclear power plants causing accidents with widespread effects. With all these events, the negative risk perception on this issue has increased. The role of the media has been decisive in the consolidation of risk perception and social representation, as exemplified in 2011 by the accident at the nuclear power plant in Fukushima, Japan, news of which was highlighted in the newspapers and was featured on the evening news for weeks. Even weeks and months later, it was kept alive in people's memory with the publication of new reports. On the other hand, any positive news regarding nuclear technology is reported on only briefly and without any highlighting or proper credit given.

It is evident that certain issues can influence public opinion and direct a social decision-making process, as discussed in Ref. [21]. This is true with regard to the RMB. From the results obtained in this study, it is clear that the communication of risks related to new nuclear facilities is an essential activity for the formation of a rational and balanced public opinion base [22]. However, as noted herein, those responsible for the communication of new enterprises still tend to rely on technical and scientific explanations to the public, which has a subjective risk perception. The role of the media in shaping the risk perception of the population is crucial [21]. It is evident that communication between the scientific community and the public is still very raw in the nuclear area.

Analysing the results presented in this article, it is clear that there is a need for intervention in the municipality for further information about the arrival of the RBM, because the population feels neither enlightened nor safe with the project. Therefore, to increase the RMB acceptance in the municipality of Iperó, an alternative may be to use the media, recognizing the socio-cultural force that it has, through interviews, articles and discussions about the issue, as discussed in Ref. [21]. The use of the media can reach a wide audience of different age groups and should be directed to it. However, it must work together with environmental education programmes and partnerships with schools, as promised at the time of the public hearings, as well as new public hearings and dissemination of information on the implementation of the RMB in schools. The public can then reflect on the topic and critically evaluate the arrival of the reactor in Iperó.

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RADIOACTIVE WASTE MANAGEMENT AND DISPOSAL

(Session 3)

Chairperson

H. MONKEN-FERNANDES
IAEA

Invited Paper

MANAGEMENT OF NORM RESIDUES: PRACTICAL ASPECTS

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Abstract

Different types of NORM residues are generated in many industries and most of them require appropriate management. The paper discusses the classification of the materials, especially the differences between 'NORM residue' and 'NORM waste' and between relevant management strategies. Several practical examples are also provided. The management of NORM residues may involve immediate removal or alternatively long term storage, prior to reprocessing. The management of NORM waste may involve long term storage prior to disposal. There are three optional strategies for disposal: (i) concentrate and contain, (ii) delay and decay, or (iii) dilute and disperse. Additional information is also provided for the dilute and disperse management option, as in many cases it is selected as the preferred one, both for NORM residues (that are blended with other materials in the process of their use) and for NORM waste (as no radiologically contaminated legacy sites are created after blended material is disposed of in mining voids and landfills).

1. INTRODUCTION

The work in this paper builds on the studies and observations by the author previously reported at TENR-2 in 1999 [1], NORM-5 in 2007 [2], NORM-6 in 2010 [3] and NORM-7 in 2013 [4], and describes the continuous developments in regard to the issues reported earlier.

NORM occurs in industries either because the activity concentration of naturally occurring radionuclides in the raw materials is higher than average, or because the activity concentration in residues and wastes is enhanced during the processing of the raw materials. There are two main mechanisms by which a radioactive residue could be generated:

(i) Large quantities of raw materials with low radioactivity are directly transformed into small quantities of residues (mass transfer), for example coal combustion;
(ii) Small amounts of radioactivity are selectively transferred from large quantities of raw materials into residues (activity transfer), for example precipitation of scales.

Typical examples of the processes of the generation of NORM residues are:

(a) Radioactive raw materials: phosphate fertilizer and titanium dioxide pigment production;
(b) Precipitation: generation of scales and sludges in oil and gas production and in water treatment;
(c) Volatilization: filter dust from coal combustion and metal smelters;
(d) Radioactive products: magnesium–thorium alloys, refractories.

2. NORM RESIDUE MANAGEMENT: PRACTICAL APPLICATION

2.1. Definitions

The first step in the management of NORM residues is their classification, in accordance with the requirements of the IAEA Basic Safety Standards (paragraph I.4 and Table I.3) and the application of the graded approach (Requirement 6) [9]. Additional information on the application of values is given in Part 5 of the IAEA Safety Guide on exclusion, exemption and clearance [6]. The next step is to define both residue and waste, in accordance with the IAEA Safety Glossary [7]:

- NORM waste: Naturally occurring radioactive material for which no further use is foreseen;
- NORM residue: Material that remains from a process and comprises or is contaminated by naturally occurring radioactive material (NORM). A NORM residue may or may not be waste.

Therefore, there is a need to establish if there is any further use for a NORM residue. It is important to bear in mind that this assessment should take into account not only current circumstances, but also future estimates. The fact that there is no use for a NORM residue at the current moment does not mean that the material has to be classified as waste. If the use of the material in the future is *foreseen*, and this fact can be proven to the satisfaction of all relevant regulatory authorities, the material could be classified as a residue and a long-term strategy for the management of its temporary storage developed.

2.2. General aspects of NORM residue management

The following general requirements apply to the management of NORM residues:

- A national policy framework within which NORM residues are managed;
- A strategy for the implementation of this policy, including the provision of the necessary resources;
- An appropriate national legal and organizational framework within which NORM residue management activities can be planned and carried out safely.

The national policy and strategy should ensure that the management system for NORM residues is consistent with management systems and requirements for residues from other industrial processes. This is very important as NORM residues commonly contain non-radioactive constituents that may be hazardous. The national policy should also ensure that the management, storage and disposal of NORM contaminated items are taken into account. A typical management framework would comprise the following:

- A Member State should determine which industries within its jurisdiction are concerned with the processing of NORM and generating NORM residues, including a *national inventory* of legacy sites, i.e. sites containing NORM residues from discontinued past practices;
- The regulatory body should have a good understanding of the technical and financial circumstances of the operator of each facility;
- The operators must have sufficient financial and human resources to enable not only safe and efficient management of NORM residues, but also a capability to manage all decommissioning and remediation activities.

It is extremely important to ensure that non-radiological contaminants that may be present in NORM residue or waste are taken into account in the development of a management strategy. The impacts of non-radiological contaminants are very often as important as radiological impacts. The understanding of non-radiological parameters may also be necessary to understand the environmental processes in the dispersion of radioactive contamination (e.g. pH, groundwater gradient). Additionally, non-radiological contaminants can be used as analogues for radioactive contaminants (e.g. natural lead for ^{210}Pb, where there is a direct relationship).

2.3. Options for the management of NORM residues

The management of NORM residues depends on when they would be used or recycled:

(1) If the residue is transferred to a use or recycling facility as it is being generated, the usual strategies for the management of the processing of a radioactive material will be applicable, as the residue generated at one facility will be considered as a ‘raw material’ at another facility.
(2) If it is expected that the NORM residue will need to be stored for a period of time before use, it will need to be placed into an authorized storage facility, sometimes requiring the development of a long-term management plan.

Typical residues generated in various industries and the possibilities for their use are listed in Table 1. Examples of the management of NORM residues are given in Section 3.

TABLE 1. SOME NORM RESIDUES AND OPTIONS FOR THEIR USE

NORM residue	Options for use or recycling
Small amounts of surface contaminated metals	Scrap metal recycling
Phosphogypsum	- Soil improvement, fertilizer - Building material - Landfill cover - Water purification - Road construction
Slag	Road construction
Mine tailings	Underground or open pit backfill
Waste rock	Construction material for tailings storage facilities and roads on mining sites
Fly ash and bottom ash	- Road construction - Inclusion in cement and concrete - Fertilizer and soil conditioner
Tailings from heavy mineral sands and bauxite processing	Open pit backfills
- Contaminated plastic, wood and rubber - Filter masses and filter cloths from water treatment, processing of titanium, rare earths, copper and other minerals - Scale and sludge from oil and gas exploration and production (including hydraulic fracturing), and from geothermal energy generation	A clear use has not yet been found

2.4. Options for the management of NORM waste

The issues with NORM waste were discussed at the very first NORM Symposium in 1997 [8]. The following approaches were proposed for the management of NORM waste and most of them are being used today: (i) shallow land or underground burial, (ii) interim storage, (iii) diluting or spreading, (iv) recycling. There are currently four options for the management of NORM residues after it has been decided that no future use for them is foreseen and they have therefore been classified as waste:

(a) Long term storage followed by disposal,
(b) 'Concentrate and contain' option,
(c) 'Delay and decay' option,
(d) 'Dilute and disperse' option.

Examples of the management of different types of NORM waste are given in Section 4.

3. MANAGEMENT OF NORM RESIDUES

In almost all known cases of the use of NORM residues the material is utilized as an additive to another product to improve its quality. It should be noted that the same blending process takes place during the dilution of NORM waste with non-radioactive material prior to disposal (Section 4.4), but in that case the aim is to ensure that there are no restrictions on the future use of the disposal site.

3.1. Immediate removal of NORM residues from a site for reprocessing

A product called silica fume (SiO_2) is generated in the production of zirconia (ZrO_2) from the mineral zircon ($ZrSiO_4$). This residue is typically generated in small quantities, of the order of several tonnes per month and, as soon as a reasonable amount has been accumulated on the site, it is transported to a customer, typically every several weeks. As noted in the IAEA Safety Report for the zircon and zirconia industries [9], during the process of fusion of zircon ^{238}U and ^{232}Th tend to stay with zirconia, but ^{226}Ra, ^{210}Pb and ^{210}Po tend to end up in the silica. As the IAEA Safety Report further states, the fine-grained silica produced in the zircon fusion process is usually sold as a by-product for use as an additive to cement and in brick making.

The concentrations of ^{226}Ra in silica fume are reported to be of the order of 5.8 Bq/g [10] and, as has been confirmed in several unpublished studies, concentrations of ^{210}Pb and ^{210}Po are typically of the same order. Therefore, care is always taken to ensure that final materials (such as cement) do not contain more than 8–10% of silica fume.

In another case, silica fume is considered to be a waste. Details are given in Section 4.4.

3.2. Long term NORM residue storage

When a NORM residue cannot be utilized in the near future, an assessment is made to ascertain if the material would be a valuable resource in the future. Two cases from Australia can serve as examples of when a use for the NORM residue has been found after a considerable amount of time.

3.2.1. Monazite concentrates

When the production of monazite in Australia ceased in the mid 1990s owing to market conditions, most of the companies processing heavy mineral sands opted for the 'dilute and disperse' option for this material, described in Section 4.4. One company, however, was able to prove to all relevant government departments that the temporary storage of monazite concentrates (containing 90–110 Bq/g of ^{232}Th and 10–15 Bq/g of ^{238}U) at the site that was classified as 'arid' and 'remote' would not cause any measurable impacts on the public and the environment. It is considered that the selected strategy was correct, as:

(a) The mineral monazite contains significant concentrations of rare earth elements — thus it is considered to be a valuable resource and some sales of monazite concentrates have already occurred;
(b) Even in the case when all accumulated material would not be sold in the intermediate future (next 20–30 years), the temporary storage location was selected in such a way that it could be converted into a long-term storage by simply covering the material in the mined out pit and having this valuable resource available to future generations.

3.2.2. Neutralized used acid

Neutralized used acid (NUA, also called synthetic gypsum) is generated in the production of synthetic rutile from the titanium mineral ilmenite and typically contains 0.5–0.7 Bq/g of ^{232}Th and 0.2–0.3 Bq/g of ^{238}U. This product is a mixture of iron oxides, hydroxides and other iron containing substances (about 30–35% in total), gypsum ($CaSO_4$) (about 60%), with minor quantities of quartz (about 5%) and manganese (about 3%). These concentration values are below those at which regulatory control may be considered in accordance with the IAEA BSS [5], but due to the fact that the product may be used in agriculture a radiological impact assessment is usually required.

This NORM residue has been stored at one of the mineral processing sites in Western Australia for several years until an application can be found — the NUA is mixed at a 5% ratio with inert sand to construct a 'nutrient filter' in order to enhance soluble phosphorus removal from surface water streams. In two years since the nutrient filter has been constructed, no changes in concentrations of NORM have been detected in the surrounding area, in water, soil and vegetation. It is expected that the application of nutrient filters could possibly be extended to other constructed wetland basins in the area.

4. MANAGEMENT OF NORM WASTE

4.1. Long term NORM waste storage followed by disposal

As described in part 3.2, in many cases NORM residues are stored for a considerable period of time before the use for them is found. However, relatively often the materials could not be utilized during the lifetime of operations or in the intermediate future and are, therefore, classified as waste requiring appropriate disposal.

4.1.1. Phosphogypsum

Very large volumes of phosphogypsum ($CaSO_4 \cdot 2H_2O$) have been generated by the phosphate industry [7, 8]. As reported by the IAEA [12], concerns about its radioactivity content and, to a lesser extent, its heavy metals content, have led to restrictions on the use of phosphogypsum in some markets, even though such concerns do not always have a proper scientific foundation. This has resulted in phosphogypsum stacks being, in effect, turned from short term holding piles into long term disposal facilities. It has been estimated that, by 2006, a total of 2.6–3.7 billion t of phosphogypsum had been accumulated in stacks worldwide.

The concentrations of radionuclides in phosphogypsum may range from 0.01 to 0.50 Bq/g of ^{238}U and from 0.02 to 3.20 Bq/g of ^{226}Ra. Therefore, in some cases specific protection measures may be required for the disposal of this material [11].

4.1.2. Tailings from the processing of titanium minerals

In the production of synthetic rutile from titanium minerals, iron is removed from ilmenite, thus increasing the titanium content from 40–60% to approximately 90% [13]. A modification of this process involves the addition of a specific flux. NORM radionuclides migrate to this flux, resulting in tailings containing NORM in concentrations that require management [13]. Radionuclides such as ^{228}Ra and ^{226}Ra become relatively mobile in the tailings from this process and must be deposited in specifically engineered lined tailings storage facilities. In almost all cases no future use for these tailings is foreseen and they are classified as waste.

At one of the locations in Australia, owing to urban encroachment the tailings dams containing these materials are now located close to residential areas, which poses a significant problem:

(i) On one hand, the likely impact of these tailings on the environment and the general public is expected to be small, and the relocation of these tailings to a mine site may be very expensive.
(ii) On the other hand, the location of a ‘radiologically contaminated site’ close to residential areas may not be acceptable from a public perception point of view and the company may have no choice but to relocate the radioactive material to the mine site, where a specifically engineered tailings disposal facility will need to be constructed.

At the moment, a final solution has not yet been found, as the company wishes to retain the tailings at their current location. However, an appropriate authority is likely to be of the opposite opinion, as the tailings are stored at the location that was not suitable even at the time when the tailings storage facility has commenced operations, in accordance with the relevant Australian Code of Practice [14] and, therefore, applicable regulations [15].

4.1.3. Other process tailings

At a medium size mineral processing plant in Asia, tailings containing approximately 9 Bq/g of ^{232}Th, 1 Bq/g of ^{238}U, 8 Bq/g of ^{22}8Ra and 2 Bq/g of ^{226}Ra are generated at a rate of approximately 90 000 t/a. When the plant was constructed, the use of this NORM residue as an additive to the road construction materials had been identified and it was stored for two years while the final testing of this use of the residue was carried out. This NORM residue was subsequently reclassified as waste and the project to use the material was abandoned, for reasons that are very important in the utilization of all NORM residues:

- Almost 1 million t of ‘clean fill’ needed to be purchased annually for blending purposes to ensure that the final mix would not require radiological management in accordance with the regulations applicable in the country;
- Even if such volume of ‘clean fill’ could have been available at a very low cost, the project was not feasible as only 15–20% of the NORM residue could have been utilized for the construction of necessary roads in this country; there was simply no need for such large volumes of road construction material in the area where the plant is located.

4.2. Concentrate and contain option

This option is utilized in cases where the volumes of NORM waste are relatively small, but the concentrations of radionuclides are relatively high.

4.2.1. Oil and gas industry

In the oil and gas industry, concentrations of ^{226}Ra can reach or exceed 15 000 Bq/g in scales inside pipes and different valves and 800 Bq/g in sludges [16]. The scales are removed at special NORM treatment plants and are typically compacted into drums that are kept in a controlled area and then disposed of at authorized facilities.

4.2.2. Titanium dioxide pigment industry

In the titanium dioxide pigment industry, radionuclide activity concentrations measured in scale and discarded filter cloths can sometimes exceed 1 500 Bq/g of ^{228}Ra and ^{228}Th [13]. The scales and filter cloths are relatively small in volume and are typically kept in dedicated sheds or containers before disposal at an authorized facility.

4.2.3. Decommissioning of NORM facilities

In the process of decommissioning of NORM facilities, one of the main aims is to ensure that valuable equipment such as pumps, tanks and conveyors are decontaminated to levels allowing for this equipment to be reused in other industries. The decontamination process always results in the generation of NORM waste — the water used for high pressure cleaning would usually contain insoluble particles. When this water is recycled through a filter a relatively small volume of NORM waste is generated, which will require appropriate disposal.

Another process in decommissioning that may result in the generation of NORM waste is the draining of processing vessels — in one of the cases in Australia, sulphuric acid was recycled over several years in the processing of a mineral containing not more than 2 Bq/g of ^{226}Ra. When the acid was drained and dried the concentrations of ^{226}Ra in the resulting residue were found to be over 2000 Bq/g.

4.3. Delay and decay option

This option is used relatively rarely, as most NORM radionuclides have very long half-lives. There are, however, two notable examples:

(1) The dust collected by electrostatic precipitators at different smelters (iron, nickel, copper) may contain significant concentrations of ^{210}Po. In cases where this dust does not contain other toxic contaminants and the concentration of ^{210}Po is the only limiting factor for the disposal of this material at an industrial landfill site, the ‘delay and decay’ approach is typically taken. For example, dust containing 300 Bq/g of ^{210}Po (half-life 140 days) will contain less than 10 Bq/g of this radionuclide after 22 months, and in just over three years the material will be exempt from radiation safety regulations in accordance with Table I.3 of the IAEA BSS [5] (activity concentration less than 1 Bq/g). Relatively often, however, this dust would also contain ^{210}Pb with a much longer half-life (22 years). In these cases, the ‘delay and decay’ option would require a long-term management plan for the material, as described in Section 4.1.

(2) A similar ‘delay and decay’ approach was taken by one of the mineral processing companies generating a waste stream with a ^{228}Ra activity concentration of 7 Bq/g (the parent radionuclide ^{232}Th reports to another tailings stream). This waste could be disposed of at an industrial landfill site when the concentration of ^{228}Ra was below 1 Bq/g, in accordance with Table I.3 of the IAEA BSS [5]. The original ^{228}Ra activity concentration of 7 Bq/g falls under the activity concentration limit of 1 Bq/g after 17 years.

4.4. Dilute and disperse option

4.4.1. Silica fume

Silica fume, described in Section 3.1, was considered to be a waste in the United Kingdom and disposed of as an industrial waste at a local landfill site until 1991, when the analysis of the material indicated that it contained 30 Bq/g of ^{226}Ra, 200 Bq/g of ^{210}Pb and 600 Bq/g of ^{210}Po [17]. Despite the fact that all environmental assessments indicated that the risks to the public and the environment were minimal, none of the disposal sites agreed to accept this NORM waste. The only possible way to dispose of this material was found to be blending it with damp sand to levels at which the material was exempt from the radiation protection regulations. Then the landfill site was not required to obtain a licence to dispose of radioactive material.

4.4.2. Monazite concentrates

Monazite concentrates, as described in Section 3.2.1, were commonly disposed of in Western Australia in mined out pits after blending with non-radioactive material, in accordance with applicable regulation [18] that states:

> "Each responsible person at a mine site must ensure that, so far as is practicable, radioactive waste is diluted with other mined material before it is finally disposed of in order to ensure that in the long term the use of the disposal site is not restricted."

Monazite concentrates containing approximately 100 Bq/g of ^{232}Th and 12 Bq/g of ^{238}U are transported to a mine site where they are blended thoroughly with mine tailings (sands and slimes, containing only trace amounts of NORM) [19]. The final tailings stream for disposal contains only 0.4–0.6 Bq/g of ^{232}Th (^{238}U is typically below the limit of detection). Therefore, no 'legacy sites' that would require institutional control for a very long period of time are created.

5. DILUTE AND DISPERSE (BLENDING) OPTION: ADDITIONAL COMMENTS

As the 'dilute and disperse' management option is considered to be the most controversial one, an additional discussion of it is warranted. Paragraph 3.29 of the IAEA Fundamental Safety Principle 7 [20] states:

> "Radioactive waste must be managed in such a way as to avoid imposing an undue burden on future generations; that is, the generations that produce the waste have to seek and apply safe, practicable and environmentally acceptable solutions for its long-term management. The generation of radioactive waste must be kept to the minimum practicable level by means of appropriate design measures and procedures, such as the recycling and reuse of material."

Therefore, the use and recycling of radioactive residues need to be considered in each case. Various IAEA publications provide additional information and recommendations with regard to the dilute and disperse waste management option. The definition of dilution is:

> "Dilution is the process in which a contaminant becomes less concentrated. It is similar for both organic and inorganic contaminants, including radionuclides. It reduces risk because resulting exposures will be lower. By itself, however, dilution does not reduce contaminant mass; rather it spreads the area of potential exposure" [21].

Observations from different IAEA publications with regard to this management method are as follows:

(a) The problem is aggravated by accumulating effects along the food chain. Another uncertainty that to date remains unresolved is the potential effect of prolonged exposure to very low concentrations. In the light of these concerns, discharges and releases have been prohibited (declared radioactive waste) or significantly curtailed in some regions of the world through international agreements [22].
(b) There is no doubt that, even where not proscribed by legislation, the dilute and disperse option is opposed by regulators, environmental groups and the public at large [23].
(c) Dilution needs to be used sensitively in order to demonstrate implementer credibility and ethics in the management of radioactive waste and thereby maintain public acceptance. Nevertheless, it is a potentially valuable technique in appropriate situations and has been used successfully [24].
(d) Some legal options for NORM residue disposal might include the release and dilution of residues into water bodies, incorporation back into the natural environment or underground placement [25].
(e) Dilution as a means of increasing the amounts of NORM residues that can be used as by-products should not only be permitted in terms of the national approach, but should actually be encouraged [26].

It is therefore clear that the dilution or blending of both NORM residues and NORM waste is the option that needs to be followed, where national regulations allow this practice. This method of management of radioactive waste is the preferred one in Western Australia [18].

A similar approach exists in the Netherlands, where the by-product use of NORM residues is the primary target of a NORM residue management system. For application in civil engineering, a specific requirement in 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 recycling or use are not feasible can the material be disposed of, and only then is it considered to be waste [27].

If national regulations allow, it is recommended that this option be followed, but the possibilities of overregulation in its application should also be considered. Unfortunately, it is not uncommon to hear an argument based on “an interpretation of an appendix of a guideline for a procedure that describes a regulation relevant to a section of an Act”. It is typically more practical to set ‘release’ or ‘use’ limits for different branches of the minerals industry and leave it to the industry itself to develop technical systems to meet these standards in specific circumstances [28].

It is very important to bear in mind that when a regulatory agency gets involved in writing detailed and compulsory specifications on how to meet the performance standards, there is a danger that the system of radiation protection will degenerate into a continuing industry effort to comply with ever more complicated regulations, procedures and guidelines, completely losing sight of the basic goal of safe operation [29].

Numerous guidelines on the use of NORM residues (providing specific ‘use limits’) have been produced at a national level. If the regulations in a particular jurisdiction do not prohibit dilution or blending, the guidelines from the European Union [30, 31], Poland [32], Finland [33], China [34, 35], Azerbaijan [36], Tajikistan [37], and Western Australia [38] can serve as useful examples. It is noteworthy that the first publication discussing the use of different coefficients for the estimation of doses and for the possible use of NORM residues in the building industry was published by the OECD in 1979, more than 35 years ago [39].

6. CONCLUSIONS

The main conclusion is that, unfortunately, there is still more confusion than certainty in the management of NORM residues and waste, and the situation has not improved significantly since 1999 [1] and 2007 [40]. Despite the best efforts of the IAEA, the Safety Reports published for the various NORM industries remain largely unknown and unused by the industries for which they were intended. A clear illustration of the issue can be drawn from the number of industry representatives attending international NORM symposia. The numbers of participants from the zircon, titanium and rare earths industries in the last three international NORM symposia were:

- NORM V, Spain, 2007: nine out of 202;
- NORM VI, Morocco, 2010: zero out of 142;
- NORM VII, China, 2013: zero out of 176.

The importance of the participation of industry in the NORM symposia is not widely appreciated, and it appears that different branches of the mining and minerals processing industry voluntarily exclude themselves from the early stages of a regulatory process that will directly affect their operations in the near future.

In addition, the following conclusions can be drawn:

(a) There are many different options for managing various types of NORM residue and NORM waste — the selection of a method would depend not only on technical and economic considerations, but also on what options are allowed by applicable local regulations, and on public opinion.
(b) A possible management method typically cannot be based on a limit of activity concentration, owing to the large diversity of NORM residues and waste and possible migration of different radionuclides into the environment. However, industry specific or material specific guidelines may be developed.
(c) Additional approvals by the relevant authority will be required in each case, based on a separate radiological impact assessment carried out for all reasonably possible scenarios.

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MANAGEMENT OF RADIOACTIVE RESIDUES AND WASTE GENERATED DURING REMEDIATION OF URANIUM PRODUCTION LEGACY SITES IN GERMANY

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Abstract

As one part of the remediation of complex legacies of uranium mining and processing operations, existing radioactive material such as ore processing waste (tailings) and host rock (waste rock) have to be safely managed. In addition, new radioactive material is generated or separated during remediation, such as when treating radioactively contaminated water or demolishing and dismantling buildings and facilities. For all this material, economic (and environmentally friendly) procedures have to be developed for by-product use, recycling or safe disposal. This paper describes the approach chosen by the German federally-owned company Wismut GmbH, which has been addressing the issue of remediating complex legacies of uranium ore mining and processing operations in eastern Germany for 25 years now. The examples considered here include the treatment of contaminated water, immobilization and disposal of water treatment residues, recycling of moderately contaminated scrap and the utilization of surplus excavated material from the remediation of waste rock piles.

1. INTRODUCTION

For more than 40 years, the former Soviet–German stock company SDAG Wismut mined and processed uranium ore in the former East Germany. By the end of 1990, the company had produced a total of 216 000 t of uranium, ranking the company as the world’s fourth largest uranium producer. Material contaminated with radionuclides of natural origin was left behind. Overriding challenges include more than 160 million m^3 of radioactive tailings from the processing of uranium ore and 300 million m^3 of moderately radioactive waste rock. The total footprint of radioactively contaminated waste rock piles, tailings facilities and plant areas amounted to 3700 ha.

Since 1991, the federally owned company Wismut GmbH has been mandated to remediate these legacies. The period of 25 years spent in remediation provides evidence of the sheer size of the legacies to be addressed and thus of the dimension of the Wismut Environmental Restoration Project. Planning for work still to be accomplished covers the period up to 2028 for physical work and up to 2045 for long term tasks such as water treatment and monitoring. Radioactive material will again be produced by these remedial efforts. Table 1 provides an overview of its volume and characteristics.

TABLE 1. CONTAMINATED MATERIAL GENERATED AND ITS RADIOLOGICAL CHARACTERISTICS

	Accumulated amount	Radiological characteristics
Seepage and pore water to be treated	17.4 million m^3 (annual volume for 2015)	– U_{nat}: 2–50 mg/L – Ra-226: 1–5 Bq/L
Water treatment residues	Uranium: 40 t (800 t in 1991) Precipitates: ~30,000 t, (500 000 t in 1991)	– U-238: 10–100 Bq/g – Ra-226: 1–20 Bq/L
Scrap from demolition and dismantling	260 000 t (cumulative total at end of cleanup)	Total alpha surface activity: 0.01–50 Bq/cm^2
Debris from demolition and dismantling	865 000 t (cumulative total at end of cleanup)	– U-238: 0.2–10 Bq/g – Ra-226: 0.2–10 Bq/g
Excavated soil from area and waste rock pile remediation	14.5 million t (cumulative total at end of clean-up)	– U-238: 0.2–10 Bq/g – Ra-226: 0.2–10 Bq/g

2. METHODS

Handling of the contaminated material has to be performed in compliance with the provisions contained in mining and radiation protection legislation and soil protection and water legislation of the Federal Republic of Germany. Furthermore, the German law on closed cycle management — the core of German waste management regulations — has to be observed. This law stipulates that residues and waste have to be recycled whenever possible. Such requirement is in accordance with IAEA standards and recommendations on the management of NORM. The principal methods for the management of solid radioactive residues and waste are:

- Use as a by-product;
- Recycling, followed by use as a by-product;
- Disposal;
- Sale.

Contaminated water has to be collected and treated prior to discharge into receiving water bodies with due regard to regulatory limits on concentrations and loads.

2.1. Contaminated seepage and minewater

Wismut operates water treatment plants at the Schlema, Königstein, Helmsdorf, Pöhla, Ronneburg and Seelingstädt sites with treatment capacities ranging from 50 to 1100 m^3/h. With the exception of the Königstein plant, lime precipitation processes tailored to site specific conditions are used to separate the radiologically relevant constituents U_{nat}, ^{226}Ra, ^{230}Th, ^{210}Pb and ^{210}Po. Because of the high uranium levels in the minewater to be treated at the Königstein site, uranium is separated prior to the subsequent lime precipitation. For the separation, an ion exchange process is applied.

2.2. Water treatment residues

Wismut sells the separated uranium from the first process stage of the Königstein water treatment plant at a break even price to a nuclear sector company which assumes responsibility for the transport and sale of the uranium on behalf of Wismut. The weighing and sale of uranium are monitored by Euratom, the competent European authority.

Sludge generated at the various sites by water treatment precipitation processes is treated with respect to its consistency (water content, particle size), chemical composition and hydrochemical and geochemical properties to render it suitable for safe disposal. In addition to the aforementioned properties, the site specific sludge treatment is also determined by technologies and requirements to be met for on-site disposal. For example, sludge at the Schlema–Alberoda site are dewatered (thickener, filter presses) and subsequently embedded within a cement based matrix. The resulting earth-moist cement solidified residue is disposed of in an engineered cell within waste rock pile No. 371 at the Schlema site and capped with a multiple layer cover. Residues are placed between layers of impermeable material and provided with drainage systems. The conceptual design and technical specifications of the engineered cell are in compliance with the requirements for a modern hazardous waste disposal facility.

In addition to residue disposal within waste rock piles, water treatment residues at Wismut are also disposed of in beach zones of tailings facilities, within an engineered cell on top of the reclaimed Lichtenberg open pit mine, as well as in specific disposal facilities for immobilized residues. With regard to their design and safety, these facilities also meet the requirements for hazardous waste disposal facilities. All disposal facilities are monitored for environmental impacts during residue placement and following closure.

2.3. Metallic scrap from demolishing and dismantling of buildings and facilities

According to a recommendation issued by the German Commission on Radiological Protection, scrap with a total alpha surface activity less then 0.05 Bq/cm^2 may be released for unrestricted use. In cases where a batch of scrap metal exhibits a total alpha surface activity exceeding 0.5 Bq/cm^2, the scrap may be recycled by melting for use in steelmaking. Scrap metal exhibiting higher levels of contamination has to be safely disposed of. In line with these requirements, Wismut has adopted the following course of action:

(1) Scrap metal classified as uncontaminated for plausibility reasons (e.g. because of its origin) is sold directly to scrap dealers.

(2) Scrap metal classified as moderately contaminated by reason of its origin is scrubbed and to some extent also decontaminated. The decontamination process uses high pressure water jetting and mechanical abrasion in a rotating container filled with small steel balls. After such treatment, the surface activity concentration of the scrap metal is measured prior to release. For that purpose, Wismut applies a screening measurement technique based on beta surface activity and a follow-up calibration of beta data versus alpha surface activity. Screening data are statistically evaluated. A batch of scrap metal may be released when the upper confidence limit of lognormally distributed data is less than 0.5 Bq/cm^2. Figure 1 illustrates a typical statistical distribution of screening data of total alpha surface activity of a batch of scrap metal. In the case under consideration, the mean of the recorded 223 measuring data is 0.14 Bq/cm^2. As the upper limit of the confidence interval (95th percentile) is 0.17 Bq/cm^2 the scrap metal may be recycled for steelmaking.

(3) Scrap metal unfit for release is deposited in engineered disposal cells within waste rock piles or in beach areas of tailings management facilities and subsequently capped.

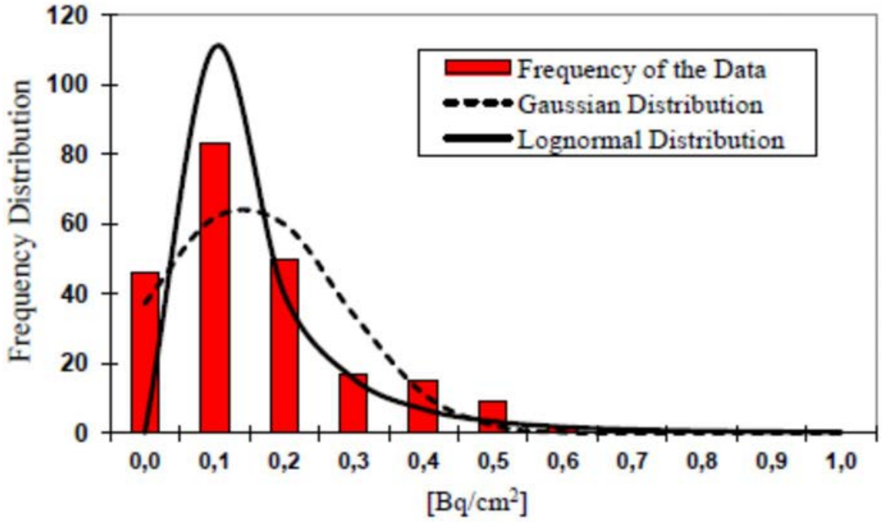

FIG. 1. Statistical distribution of total alpha surface activity of a batch of scrap metal.

2.4 Debris from the demolition of buildings and facilities

In accordance with mining legislation, uncontaminated demolition debris must not be delivered to disposal sites operated by Wismut. It has to be deposited at municipal landfill sites. Contaminated demolition debris is deposited in engineered disposal cells within waste rock piles or in beach areas of tailings management facilities and subsequently capped. The disposal cells are partly sealed at the bottom by a compacted low permeability soil layer. The purpose of the bottom seal and the overlying cap is to ensure that water infiltration and seepage-borne release of contaminants into the environment are as low as reasonably achievable.

2.5 Surplus excavated soil from the remediation of industrial areas and waste rock piles

Highly contaminated excavated soil is treated similarly to contaminated demolition debris and deposited in engineered disposal cells within waste rock piles or in beach areas of tailings management facilities and subsequently capped. For moderately contaminated excavated soil and surplus excavated materials from waste rock pile remediation, the Wismut Environmental Restoration Project provides two options for use as a by-product:

(a) Placement into interim covers of large scale tailings management facilities for regrading and contouring of the tailings surface to enhance precipitation runoff from the final cover;
(b) Use for refilling of surface subsidence and for the stabilization of slopes and dams.

3. RESULTS

Wismut's management of radioactive residues and waste originating from remediation work has contributed to the gradual decline in environmental impacts at the sites under remediation. By way of example, the introduction of efficient water treatment processes has diminished annual uranium releases to surface water bodies from 27 t originally to less than 1.9 t at present.

Residues and waste that fail to meet the requirements for by-product use or recycling are put into safe long term storage. In accordance with legal provisions, their environmental impacts have to be reduced by technical measures to a reasonably low level.

Release of scrap metal for recycling has not only a positive environmental impact; it is also a success from an economic point of view. In line with the current trend, more than half of the estimated volume of 260 000 t of scrap metal originating from or still being generated by the Wismut Project is anticipated to be released for by-product use, recycling or sale.

4. DISCUSSION AND CONCLUSIONS

Environmentally responsible and economic management of residues and waste generated during the remediation of complex uranium mining and processing legacies poses a challenge to national authorities and operators associated with large scale remedial projects. Development and implementation of technologies have to comply with conditions specific to the site. At the same time, national laws and regulations have also to be observed.

In Germany, operators have to take into consideration the requirement for by-product use or recycling of radioactive material generated during remediation. However, German mining legislation, as well as radiation protection legislation applicable to the Wismut project, imposes restrictions on the by-product use of contaminated material. Material with an activity concentration of the dominant radionuclide (usually ^{226}Ra) exceeding 0.2 Bq/g is regarded as radioactive. Waste rock, most of which has an activity concentration less than 1 Bq/g must not be used outside the Wismut property, for example as material for road foundation purposes. This is in contrast to more recent approaches to the management of NORM, as also recommended by the IAEA.

Nevertheless, the Wismut case study provides ample basis for sharing experience on the management of large amounts of radioactive residues and waste. In the meantime, remedial technologies and approaches developed by Wismut are being applied to uranium mining remedial actions at sites outside of Wismut.

RADIONUCLIDE BEHAVIOUR IN POTENTIAL APPLICATIONS OF PHOSPHOGYPSUM

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Abstract

Phosphogypsum was used alone and in cement mixtures for investigating the behaviour and fate of radionuclides contained in radioactive phosphogypsum. As production of cement with incorporation of phosphogypsum and later use of this cement in construction will involve steps with heating to very high temperature, contact with water, and exposure to air and weathering, these parameters may constrain the release of radionuclides such as isotopes of U, Th, Ra, Rn, Po, Pb in the cement life cycle. Results indicate that clinker production may largely volatilize polonium and radium from the cement but not uranium and thorium, and contact with water will dissolve radium from the phosphogypsum. Radon emanation from concrete structures using this cement will be low. Long term leaching of radionuclides from concrete structures by rain or other contact water is under investigation.

1. INTRODUCTION

There is a large worldwide legacy of phosphogypsum piles, estimated at about 3×10^9 t, located in countries producing phosphoric acid from phosphate rock. Attempts have been made to use phosphogypsum, such as in plaster for civil construction, as soil amendment, as road construction material, in brick fabrication, and as a component of cements. A major drawback in such applications of phosphogypsum is the radioactivity very often present at high concentrations in this material [1, 2]. To avoid increasing the exposure of members of the public to additional radiation doses, any use of phosphogypsum containing non-negligible concentrations of radionuclides has to go through thorough assessment of the radiological risk associated with that use. The radiological risk depends on radionuclide concentrations but largely depends also on the mobility of radionuclides from phosphogypsum and scope for generating radiation exposure and radionuclide transfer to human beings.

Among the several applications attempted, the incorporation of radioactive phosphogypsum in cement (and thus concrete) could possibility better immobilize radionuclides and prevent their dispersal in the environment. Cements usually contain natural gypsum, up to 5 wt%, added during production of the clinker in order to ensure water retention and provide suitable binding characteristics to the concrete. We tested partial and total replacement of natural gypsum (which contains low radioactivity) by radioactive phosphogypsum in the cement manufacture. In the production process, the main raw materials limestone and marga rock are pulverized, then heated to a high temperature (above 1000ºC) to eliminate carbonates and obtain the clinker. Often, other materials are incorporated as well, such as flyash from coal power plants, and pozzolanas. The opportunity of using the cheaper and abundant phosphogypsum instead of natural gypsum in cements may largely depend on the fate of radionuclides contained in the phosphogypsum during cement manufacture and, later, radionuclide behaviour in the matrix of concrete structures built with that cement.

Several experiments were performed to understand the behaviour and fate of radionuclides initially contained in phosphogypsum during cement fabrication, including heating to high temperatures to produce clinker and prolonged contact of concrete blocks with water.

2. MATERIALS AND METHODS

A set of analyses and tests was planned to evaluate the initial radionuclide concentrations in the phosphogypsum and in other components of the cement, in order to assess radioactivity incorporated by the addition of phosphogypsum, the volatilization of radionuclides from the phosphogypsum during the intense heating to produce clinker, the solubility of radionuclides from phosphogypsum in contact with water, radon emanation from cement blocks built with cement containing phosphogypsum, and the long term leaching of radionuclides from concrete blocks containing phosphogypsum.

Cement materials and cement mixtures were prepared in the laboratory of a cement factory. Analyses of radionuclides were made by alpha spectrometry and validated radiochemical procedures [2, 3]. In brief, after accurate weighing, the sample aliquots were transferred to teflon beakers and accurately known activities of radioisotopes for use as internal tracers (^{232}U, ^{229}Th, ^{224}Ra, ^{209}Po, 10 mg Pb) were added to the beakers. Thereafter, the samples were covered with concentrated HNO_3, HCl and HF and maintained under controlled heating in a microwave sample digestor, until complete dissolution of the solids. After elimination of HF by further heating, the sample residues were dissolved in HNO_3 and transferred to ion chromatography columns to separate U, Th, Ra, Pb and Po radioelements. After separation and purification of these fractions, radionuclides were electroplated either on stainless steel discs or silver discs. These metal discs were used as alpha sources for the analysis of alpha particles emitted using silicium detectors and OCTECTE Plus (Ortec EG&G) spectrometers.

3. RESULTS AND DISCUSSION

Results of analysis from several experimental tests are presented herein. Table 1 shows some examples of radionuclide concentrations in cement components. Sandstone, silica sand, and natural gypsum all contain natural radionuclides in concentrations much lower than in the phosphogypsum. It should be noted that radionuclides in other cement additives, such as flyash, may be relatively elevated as well.

TABLE 1. RADIONUCLIDE CONCENTRATIONS IN PHOSPHOGYPSUM AND IN COMMON COMPONENTS OF CEMENT

	Radionuclide concentration (Bq/kg)				
	^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po
Phosphogypsum	197	727	345	827	827
Components of cement:					
Limestone	24	28	24	17	32
Process sand	19	29	33	16	15
Flyash	107	102	855	110	99
Natural gypsum	21	17	30	15	21
Clinker	38	55	30	42	6

Note: Propagated analytical uncertainties were around 5–10% of values.

Where phosphogypsum was added during the production of clinker, some of the radionuclides incorporated with the phosphogypsum were released by volatilization. After heating at 1000ºC, no more than 37% of the ^{226}Ra and 5% of the ^{210}Po remained in the clinker. Nevertheless, uranium and thorium isotopes were not volatilized and remained in the clinker, being transferred to the concrete, as illustrated in Fig. 1. A concrete structure built with this cement will have a low radium content and thus will be a reduced source of ^{222}Rn emanation compared with phosphogypsum.

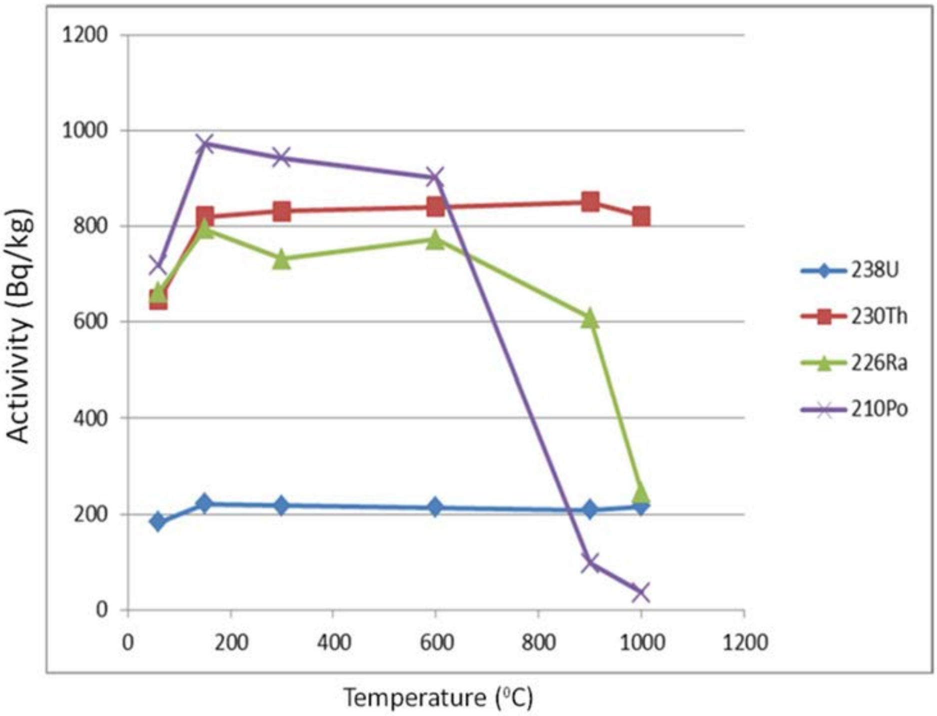

FIG. 1. Evolution of radionuclide concentration with calcination temperature of phosphogypsum.

Contact of phosphogypsum with water before and after cement application may dissolve some of the radionuclides contained therein. Tests carried out with distilled water, brackish water and seawater showed that only about 1% of thorium is dissolved, and only about 3% of polonium and uranium did get into solution. Radium was much more soluble and its dissolution increased from distilled water, to brackish water to seawater where 87% of radium initially contained in phosphogypsum became dissolved (Fig. 2). Water insoluble radionuclides remained in the cement matrix.

Phosphogypsum in cement may release radionuclides in contact with water. Nevertheless, after application of the cement, the concrete hardens in hours to days and becomes less exposed to contact with water. From the hardened concrete, radionuclides can only be extracted from reduced surface areas and by pore water from inside the structure. In the long run, radium as the more water soluble radionuclide might be the most problematic one. Long term tests (3 years) on radionuclide leaching from concrete blocks are underway to clarify this behaviour.

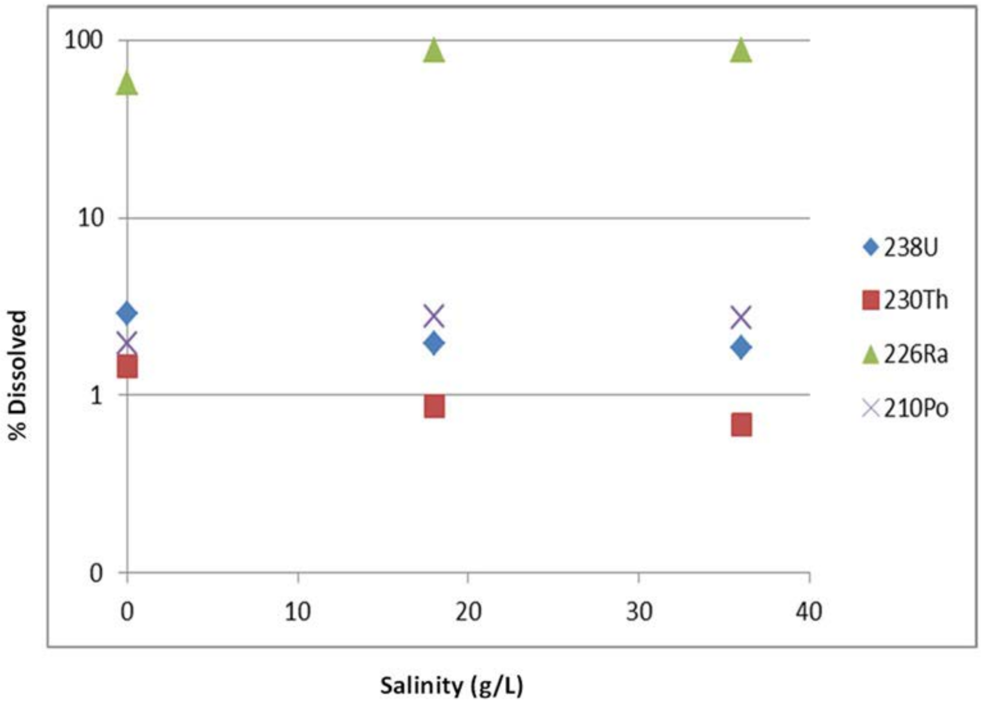

FIG. 2. Dissolution of radionuclides from phosphogypsum with increased salinity of water.

4. CONCLUSIONS

The use of phosphogypsum containing radionuclides in high concentrations may give rise to additional exposure of the public to ionizing radiation, as recognized in many circumstances [4–6]. Careful assessment of radiation doses, and thus the behaviour of radionuclides contained therein should be performed.The use of phosphogypsum as a cement component may submit radionuclides to physical–chemical conditions that play an important role in their fate.

In a cement factory, the strong heating of phosphogypsum during clinker production may significantly decrease radioactivity in the cement through volatilizing some radionuclides (Ra, Po) but, at the same time, this would generate significant releases of ^{226}Ra and ^{210}Po that may remain in the furnace or escape to the atmosphere. This will require a throrough radiological risk assessment.

The heating will immobilize the non-volatilized radium and other radionuclides (U, Th) in the cement matrix, rendering them insoluble to concrete pore water later. If this is the case, the incorporation of radioactive phosphogypsum in cement aimed to be used in outdoor constructions, such as viaducts and bridges, may present a very small, and eventually a negligible, radiological risk.

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NORM MANAGEMENT IN THE NETHERLANDS

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Abstract

The Netherlands has a long history of dealing with NORM, starting at the end of the 1970s and the beginning of the 1980s. As a result of this early involvement, the industry, regulatory bodies and radiation protection community (Dutch Health Physics Society) have overcome major challenges that are inevitably linked to NORM, due to, for example, the relatively low activity concentrations and large quantities and volumes in comparison with waste containing artificial radionuclides, and to particular issues associated with the use or disposal of residues. Procedures for working with NORM in the industry as well as associated regulations have been gradually developed through experience gained and best practice. These procedures and regulations can be considered as being among the most mature and well established worldwide. This gradual approach, including lessons learned, are described and discussed in this paper. It is evident that systems have to be altered to fit future (sustainable) insights, regulations, etc. Therefore in this paper additional information is presented about:

- Historical and existing amounts of regulated NORM materials as well an assessment of future amounts;
- The occurrence of NORM that has to be dealt with;
- Regulatory classification and registration of NORM practices;
- Radiation protection measurements in the workplace;
- NORM predisposal management;
- Available disposal options for the various NORM residues;
- Challenges still being faced, such as the implementation of new European legislation and exposure of the public to radon and thoron in homes.

1. THE PROCESS FROM NORM AWAKENING TO MATURITY IN THE NETHERLANDS

This development from NORM awakening to maturity in the Netherlands has been gradually achieved over the past few decades. A summary of this development is presented below and is based on available literature.[1] This process can be roughly divided into six time periods:

(i) Awakening period (1975–1985),
(ii) Basic document on radon (1985–1991),
(iii) European Council Directive 96/29/Euratom (1991–1996),
(iv) Implementation of Directive 96/29/Euratom through national legislation (1996-2002),
(v) Implementation of national legislation into practice (2002–2011),
(vi) Evaluation and the future (2011 onwards).

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[1] The summary is intended only to outline the process, and not necessarily to be complete.

1.1. Awakening period (1975–1985)

The awakening started in 1975, when preliminary results were reported about the elevated ^{226}Ra activity concentration in coal and flyash [1]. These preliminary results triggered a number of additional investigations:

(a) A more thorough study of elevated levels of uranium in flyash in comparison with the original coal [2];
(b) A survey initiated by the Dutch regulator with its main aim being to "collect data to be able to judge on the radiation dose caused by construction materials" [3];
(c) A study on "Radioecology of and radiation dose from Dutch waste gypsum released into the environment" [4].

This awakening period can be best described as the period that health physicists, regulatory bodies, etc. became aware of NORM and its possible contribution to the collective and individual dose. Looking at the regulations at that time, it can be concluded that most regulations were developed for the nuclear industry and did not take into account radiation effects caused by natural materials.

1.2. Basic document on radon (1985–1991)

The regulatory body initiated a study to determine (i) the radiological risks for man and the environment with special attention to indoor conditions in homes, (ii) the technical possibilities for reducing these risks, and (iii) potential policy scenarios in relation to radon [5]. Based on this study it was advised that the focus should be on measures in newly built houses as this would lead in time to a substantial reduction of the individual risk. In 1988 a study on the radiological properties of consumer goods was finalized. It was estimated that consumer goods were responsible for 90% of the collective dose [6].

In that period much effort had also been dedicated to improved sampling and gamma spectroscopic techniques for NORM [7]. The reduction and control of the risk to the public due to radon were addressed in more detail by the regulatory body in a national policy document. At that time, the regulatory body was in principle ready to formulate a policy and/or law to regulate exposure of the public to NORM as far as dwellings (and, to some extent, industry) were concerned. However, the consequences of such a policy and/or law on the industry were still not completely understood. Therefore the Minister decided to apply the 'stand still principle'. This was the start of more than a decade of research and discussions between the stakeholders. In this period also, a first outline of NORM related problems in industry became apparent.

1.3. European Council Directive 96/29/Euratom (1991–1996)

The milestone in this third period was the development of European Council Directive 96/29/Euratom for the health protection of the general public against the danger of ionizing radiation [8]. This Directive includes a section dealing with the exposure of the public and workers to natural sources. It was clear that the choice of values of clearance and exemption levels for naturally occurring radionuclides could have a large impact on the industry. Therefore national and international studies were initiated by the regulatory body and by private companies to assess the problem [9], as well as to determine the radiological risk associated with NORM and the consequences of possible clearance and exemption levels for different industries [12–15]. The effect of these studies was that other countries also became aware of the problems that could be faced by having criteria for NORM that were too strict. It is believed that the active approach by the Dutch regulatory bodies, both at a national and international level, positively influenced the industry in establishing exemption and clearance levels for NORM in Directive 96/29/Euratom.

1.4. Implementation of Directive 96/29/Euratom through national legislation (1996–2002)

During preparation for the implementation of Directive 96/29/Euratom through national legislation, the government and the industry initiated supporting studies [16–19]. As one of the final steps in this preparation, the government initiated an evaluation of the basis for the proposed clearance levels to be applied in the new national legislation [20]. It should be noted that during this period a close and fruitful cooperation was established between regulatory bodies, industry and other stakeholders. In 2002, the new Dutch legislation based on Directive 96/29/Euratom entered into force.

1.5. Implementation of national legislation into practice (2002–2011)

In 2002, the results of a second study entitled Radioactivity in Dutch Consumer Articles were published [21]. One of the conclusions of this study was that the annual collective dose had dropped from 130 manSv in 1988 to 4.6 manSv in 2002. This decrease was mainly caused by the replacement of NORM containing materials by non-radioactive materials. However, new production and construction methods, as well as ventilation techniques aimed at energy efficiency, caused an increase in the average radon concentration in homes. Levels increased from an average of 20 Bq/m^3 in 1970 to an average of 30 Bq/m^3 in 1990 [22].[2]

Specific legislation for NORM in the Netherlands started in 2004 with an ordinance concerning natural activity of sources of ionizing radiation (the NABIS Ordinance) [23]. This was revised in 2008. As a result, the average dose due to radon slowly decreased at an estimated rate of 1.5 Bq/m^3 per decade [24].

Nowadays, different treatment routes are available for different kinds of NORM residues, e.g. use, landfill disposal, recycling by melting (metals) and, as a last option, interim storage [25]. The appropriate route is determined by legislation, infrastructure and economics. The use of NORM residues is explicitly mentioned in the NABIS Ordinance, as is the possibility of disposing hazardous NORM waste in a specified waste repository (a landfill site).

By restricting the use of NORM in consumer products, collective doses are decreasing and some of the minerals processing industries are starting to take into account the radiological properties of their process materials along with other parameters. It can be concluded, therefore, that the regulatory policy is starting to be successful.

1.6. Evaluation and the future (2011 onwards)

A new, revised Directive on radiation protection (Directive 2013/59/Euratom) was adopted by the Council of the European Union in 2013. Its implementation in the Netherlands will necessitate considerable changes to the NABIS Ordinance. It is expected that studies will have to be carried out to investigate the consequences of the application of NORM by-products (in the construction and maintenance phases) as well as the consequences of NORM releases during the demolition of structures and the possible (secondary) use of materials.

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[2] Based on improved radon measurements, the author of Ref. [22] reported also that the average radon concentrations reported in the past might have been influenced by the exhalation of thoron. The specifics of thoron detection and assessments of its influences are beyond the scope of this paper.

The Dutch legislation for NORM has been compared with that in neighbouring countries [26]. The findings are that the Dutch legislation is relatively strict and, as a consequence, the administrative costs are relatively high. Some existing rules are rather complex and unclear and give rise to problems with the enforcement of the legislation. This outcome has motivated the start of discussions with stakeholders about relaxation of legislation. The regulatory body has pointed out that there is room for improvement in national legislation in terms of: (i) clarification of existing rules and regulations, (ii) a multi-stage risk assessment approach (iii) relaxation of releases into soil as a result of gas production. The regulatory body has also expressed a need to act upon these findings at an international level, for instance by proposing modifications to the European Directive [26].

1.7. Discussion

The above summary describes 'the process from NORM awakening to maturity in the Netherlands' based on facts and milestones of this process reported in the literature. It is evident that, during this process (already covering a time span of almost 50 years), a large number of organizations and people have been participating. It should be mentioned that there have been no serious drawbacks during this process and even the current stakeholders in the process are still cooperative and have confidence in each other and in the ongoing process.

2. HISTORICAL, EXISTING AND FUTURE AMOUNTS OF REGULATED NORM

The total amount of radioactive waste in the Netherlands is some hundreds of thousands of cubic metres. The majority of this comprises low level radioactive waste and NORM residues, which are stored at various waste repositories such as landfill sites. The quantity of all radioactive waste and NORM residues currently stored at the Central Organization for Radioactive Waste (COVRA) is about 35 000 m^3. Of this, more than half (20 500 m^3) is NORM, which in turn comprises 14 000 m^3 of depleted U_3O_8 and 6 500 m^3 originating from other NORM industries. Extra (dedicated) storage capacity for depleted U_3O_8 is presently under construction. The NORM waste stored at COVRA contains about 0.02% of the total activity stored at that facility [27].

As with other types of waste, the laws and regulations (and developments thereof) are of great importance for the definition of radioactive waste. Following recent changes in the definition of radioactive waste, the volumes to deal with have increased from several hundreds to a few tens of thousands of cubic metres per year. Much of this waste is in the 'notification obligation' range (see Section 4) and is thus close to the current exemption levels. There are also quantities of waste that are below the current exemption levels but which nevertheless have elevated radionuclide concentrations. This means that even small changes in the exemption levels may bring about major changes in the total amount of NORM waste.

Figure 1 shows the estimated growth in the quantity of NORM stored at COVRA until the year 2130. However, should the use of NORM residues or their disposal at licensed landfill sites be no longer permitted, the amount would increase at a rate of nearly 40 000 m^3 per year and the estimated quantity of NORM stored at COVRA in 2130 would increase from about 150 000 m^3 to 3.8 million m^3.

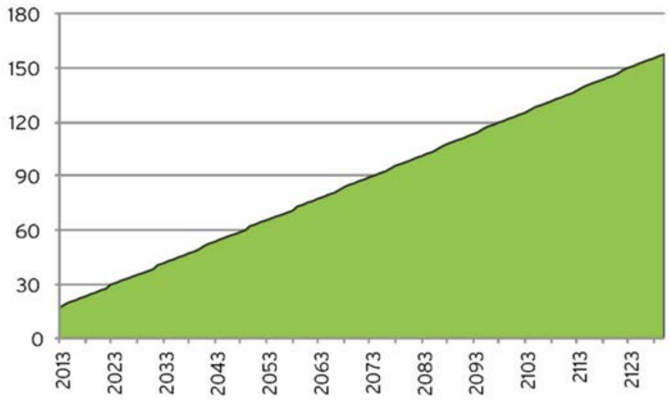

FIG. 1. Estimated quantity of NORM (in thousands of cubic metres) stored at COVRA in future years.

3. OCCURRENCE OF NORM

The Netherlands is similar to other countries in that it has a large variety of industries that generate NORM. The main industries and examples of the types of NORM are as follows:

- Steelworks (slag),
- Coal fired power plants (flyash),
- Uranium enrichment facilities (depleted U_3O_8),
- Oil and gas production (contaminated equipment and structures, sludge, scale),
- Pigment production (residues),
- Fertilizer production (contaminated equipment and structures),
- Phosphorus industry (contaminated equipment and structures),
- Metal scrap (thorium contaminated metal casings with insulation wool),
- 16 other industries.

An overview of the NORM industry in the Netherlands is presented in Fig. 2.

FIG. 2. Overview of the NORM industry in the Netherlands.

4. REGULATORY CLASSIFICATION AND REGISTRATION OF NORM PRACTICES

The Nuclear Energy Act is the basis of Dutch regulations in the field of nuclear safety and radiation protection, and thus for the management of spent fuel, NORM and radioactive waste. In addition to the Nuclear Energy Act are some other laws relevant to the management of spent fuel and radioactive waste as the ‘Radiation Protection Decree’. These laws cover, among other things, liability in the case of accidents with nuclear facilities, the responsibilities of supervisors, open government, public participation and legal protection [28]. In the Nuclear Energy Act, levels are given for exemption and clearance of radioactive materials. These levels are based on Directive 96/26/Euratom. In the Netherlands, the choice has been made to use the same levels for exemption and clearance. However, for work activities up to a tenfold higher level, a notification procedure is required instead of a licensing procedure. This procedure is laid down in the Ministerial Regulation, Natural Sources of Ionizing Radiation, 2008 [23].

5. RADIATION PROTECTION MEASUREMENTS IN THE WORKPLACE

Following the adoption of Directive 96/26/Euratom, the regulator initiated two studies, reported in 1999, on the following topics:

(i) Identification of activities with exposure to natural radiation sources,
(ii) Work with exposure to natural radiation sources.

Based on these studies a classification of work has been set up according to the dose received by workers. To assist the industry, the regulatory body has published a list of activities in which the industry had to determine whether notification or permit limits will be exceeded. The annual dose criteria for employers are 0.1 mSv for normal work and 1 mSv for ‘unfavourable work.’ For members of the public the dose criteria are 1 mSv ambient dose equivalent and 0.3 mSv effective dose. The industry had to notify the regulatory body about the outcome of its assessments.

6. NORM PREDISPOSAL MANAGEMENT

Management of NORM residues in the Netherlands is based on the following hierarchical approach (the ‘ladder of Lansink’):

(i) Prevention, e.g. utilization of natural resources with lower levels of radioactivity, process adjustments to eliminate or minimize the amount of NORM residue that has to be disposed of as waste;
(ii) Use, e.g. decontamination of equipment and structures;
(iii) Recycling e.g. contaminated scrap metal to smelters, use of process residues in other industrial processes and products such as the use of slag for construction of roads and dikes;
(iv) Incineration to bring about volume reduction;
(v) Disposal as waste, e.g. notifiable waste sent to landfill sites and licensable waste sent to COVRA.

It should be noted that the regulatory body is in favour of the use of NORM residues as much as possible. This is expressed and supported by a Ministerial Regulation [23] in which it is stated that the mixing of material with elevated NORM levels is allowed for purposes of decreasing these levels and thus maximizing the use of such materials.

7. AVAILABLE DISPOSAL OPTIONS FOR VARIOUS TYPES OF NORM

In accordance with Article 37 of the Dutch Radiation Protection Decree:

(a) Notifiable waste is allowed to be disposed of at a type C2 landfill site for the disposal of hazardous and toxic waste with an additional licence for the disposal of notifiable waste. This means that notifiable waste also has to meet the regulatory requirements for hazardous and toxic waste. Notifiable waste not meeting such regulatory requirements has to be disposed of at COVRA.
(b) Licensable radioactive waste, regardless of whether it is NORM waste or not, has to be disposed of at COVRA.

In terms of Article 37, there is in principle always a disposal option available for notifiable and licensable NORM waste. Clearly, however, the associated costs will differ between the two disposal options.

8. CHALLENGES STILL BEING FACED

The following challenges continue to be faced in the Netherlands:

(a) Implementation of Directive 2013/59/Euratom (mentioned already in Section 1.6): The regulator has already embarked upon this process in consultation with stakeholders, having due regard for the potential cost implications for industry.
(b) Measures to decrease the exposure of members of the public: The annual dose received by a member of the public is, on average, 2.5 mSv, about 40% of which is due to the inhalation of radon and thoron and to exposure to radionuclides in construction materials; in homes, about 70% of exposure is due to radon.
(c) Investigation of the contribution of thoron to indoor exposure.
(d) Development of characterization techniques for NORM that are rapid (e.g. 30 min) and easy to operate: Such techniques will be needed in order to implement legislation based on Directive 2013/59/Euratom.
(e) Development of disposal options for ‘relatively small’ amounts of hazardous and toxic notifiable NORM waste, so that this waste can be disposed of at a landfill site instead of at COVRA.
(f) Development of regulations to ensure that, in the event of a major NORM industrial enterprise becoming insolvent, the government does not have to assume legal and financial responsibility for decontamination and environmental remediation of the site.

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RADIOLOGICAL RISKS ASSOCIATED WITH BUILDING MATERIALS AND INDUSTRIAL BY-PRODUCTS

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Abstract

To get a better insight into the radiological features of natural raw materials and industrial by-products that can be used in building materials, a review of the reported scientific data can be very useful. The current study is based on the continuously growing database of the By-BM (H2020-MSCA-IF-2015) project (By-products for Building Materials). Currently, the By-BM database contains individual data on 1095 raw materials and 431 industrial by-products used for building. It was found that radionuclide concentrations in the raw materials varied widely — from less than detection levels up to 27 851 Bq/kg for 226Ra, 906 Bq/kg for 232Th and 17 922 Bq/kg for 40K — while the radionuclide content of the by-products varied less widely — from 7 to 3152 Bq/kg for 226Ra and from less than the detection limit up to 1350 and 3001 Bq/kg for 232Th and 40K, respectively. The average 226Ra, 232Th and 40K concentrations in the industrial by-products were, respectively, 2.52, 2.35 and 0.39 times those in the building materials. Gamma exposure arising from bulk building products was calculated (i) using the 'I-index' approach described in the European Commission publication Radiation Protection 112, based on a single, fixed value for the material density and (ii) as described in IAEA Safety Standards Series No. SSG-32 using measured values of material density. It was found that in most cases the I-index approach (without consideration of the material density) resulted in a significant overestimation of effective dose.

1. INTRODUCTION

Radiation exposures due to minerals containing radionuclides of natural, terrestrial origin (^{238}U decay series, ^{232}Th decay series and ^{40}K) are generally not significantly higher than normal background levels. Average worldwide activity concentrations are currently 33, 32, 45 and 412 Bq/kg for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, respectively [1]. In the case of building materials, average worldwide values are 50 Bq/kg for ^{226}Ra, 50 Bq/kg for ^{232}Th and 500 Bq/kg for ^{40}K [2]. Although, these average activity concentrations are relatively low, large variations can be found and, as a result of the occurrence of anomalies, elevated concentrations can be found in some natural materials. In many cases, these materials with elevated concentrations have been used as building material. The determination of the radionuclide content of building materials and assessment of indoor exposure is therefore important because most individuals spend 80% or more of their time indoors. Chronic exposure involving moderate doses of radiation can increase the risk of health damage to individuals, which may occur decades after the exposure [3].

Building materials can be produced directly from natural materials such as rocks, granite and clay or from industrial by-products such as flyash, bottom ash, steel slag and bauxite tailings ('red mud'). It is important to investigate the use of such materials in order to allow them to be safely and efficiently integrated into new and refurbished buildings. To get an insight into the radiological features of potentially usable industrial by-products, a review of the reported scientific data is necessary. This study is based on the continuously growing database of the By-BM (H2020-MSCA-IF-2015) project. The aim of this project is to characterize the mechanical and radiological parameters of constituents and prepared By-BM geopolymers made from industrial by-products. This project is connected to, and provides information for, the NORM database COST TU 1301 NORM4Building Action.

2. MATERIALS AND METHODS

In order to draw conclusions from the scientific data embedded into the By-BM database, unified selection criteria were laid down:

(a) To establish an overview of the scientific data, individually reported sample data for ^{226}Ra, ^{232}Th and ^{40}K obtained by gamma spectrometry were used — the ranges of activity concentrations were not usually imported into the database. After extracting the data, criteria were applied to ensure a meaningful statistical analysis.
(b) Average concentration values for certain materials were used only if the investigated material originated from the same site, e.g. a quarry, mine or reservoir. In the case of commercial building material, the brand and type of each sample had to be clearly mentioned in the reference to fulfil selection criteria. Furthermore, the range of the data was also checked and the mean was used only if the minimum and maximum values were within 20% of the mean.
(c) ^{238}U activity concentrations were imported into the database only if the results were obtained from the measurement of the concentrations of ^{222}Rn progeny (^{214}Bi and ^{214}Pb) to avoid errors due to decay chain disequilibrium.

The so-called ‘I-index’ is widely used for evaluating building materials with respect to gamma exposure [4]. The index is calculated using equation (1).

$$I = \frac{C_{\text{Ra}-226}}{300} + \frac{C_{\text{Th}-232}}{200} + \frac{C_{\text{K}-40}}{3000} \qquad (1)$$

where $C_{\text{Ra-226}}$, $C_{\text{Th-232}}$ and $C_{\text{K-40}}$ are, respectively, the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in becquerels per kilogram. This calculation method is based on a model described in Ref. [5] for a building constructed from concrete with a density of 2350 kg/m^3 and a wall thickness of 20 cm. An I-index of 1 is specified in Ref. [6] as a conservative screening criterion for identifying materials that, when incorporated into a building in bulk quantities, might give rise to an individual dose of more than 1 mSv above background. In terms of Ref. [6], the dilution and mixing of construction materials is permitted as long as the index of the final building product itself is below a value of 1, which makes possible the mixing of by-products with low activity level raw materials.

In order to determine the dose more precisely, the actual density and thickness of the material (rather than the fixed values specified above), as well as factors relating to the type of building and the intended use of the material (bulk or superficial) need to be taken into account. In Ref. [7], the dose modelling used in Ref. [4] is again applied, but this time using the actual density and thickness of the building material.

3. RESULTS AND DISCUSSION

The current version of the By-BM database contains data on 1095 natural materials used for building and 431 industrial by-products, gathered from 48 countries. The worldwide distribution of data sources and information on material density and type are shown in Fig. 1 and Tables 1 and 2.

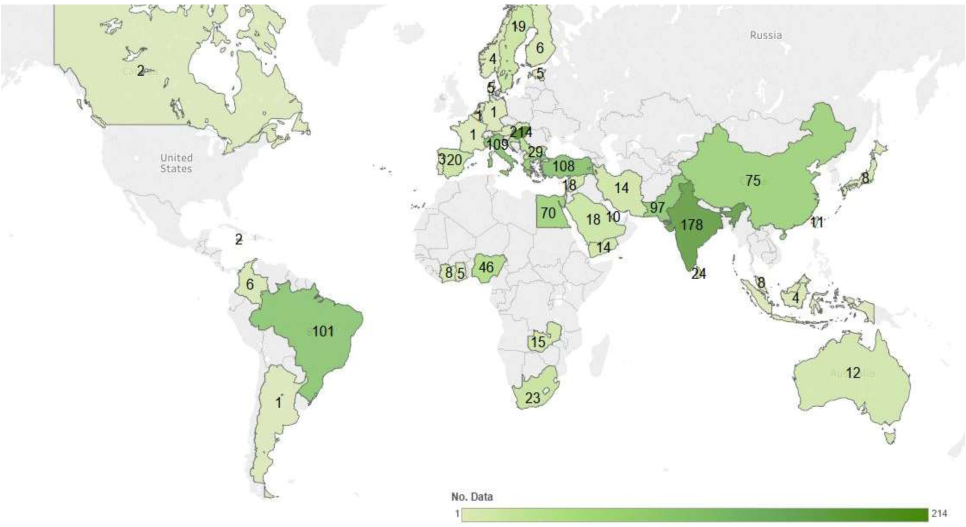

FIG. 1. Worldwide distribution of data sources for building materials.

TABLE 1. RAW MATERIALS USED FOR BUILDING

	Number of data sources	Density (kg/m^3)
Aggregate	9	1900
Basalt	3	3000
Brick	243	1900
Cement	87	1500
Ceramics	94	2400
Concrete	63	2350
Cellular lightweight concrete	37	700
Granite	297	2600
Gypsum	66	865
Limestone	16	2600
Marble	72	2550
Pumice	3	650
Rock	31	2300
Sand	19	1500
Sandstone	14	2323
Serizzo	5	2650
Sienite	5	2700
Asbestos tiles	4	1750
Travertine	9	2300
Tuff	10	2100
Volcanic	7	1800

TABLE 2. INDUSTRIAL BY-PRODUCTS

	Number of data sources	Density (kg/m^3)
Bottom ash	59	700
Flyash	145	720
Manganese clay	44	2800
Phosphogypsum	45	1500
Bauxite tailings (‘red mud’)	92	1600
Steel slag	41	2600
Titanium dioxide production residue	5	4300

Individual data on ^{226}Ra, ^{232}Th and ^{40}K activity concentrations are available for 30 different materials (23 raw materials used for building and 7 industrial by-products). In the case of the raw materials used for building, it was found that radionuclide concentrations varied widely — from less than detection levels up to 27 851Bq/kg for ^{226}Ra, 906 Bq/kg for ^{232}Th and 17 922 Bq/kg for ^{40}K — while the radionuclide content of the industrial by-products varied less widely — from 7 to 3152 Bq/kg for ^{226}Ra and from less than the detection limit up to 1350 and 3001 Bq/kg for ^{232}Th and ^{40}K, respectively. The average ^{226}Ra, ^{232}Th and ^{40}K concentrations in the industrial by-products were, respectively, 2.52, 2.35 and 0.39 times those in the raw materials — this illustrates why, when considering possible radiation exposure, the radionuclide content of such by-products generally cannot be ignored.

In this study, the absorbed gamma dose rate received by an individual was calculated using a model room with a 20 cm wall thickness and with various densities of material, using activity concentration data for ^{226}Ra, ^{232}Th and ^{40}K obtained from the database. The dose calculation was carried out using (i) a fixed density of 2350 kg/m^3 in accordance with the modelling method adopted in Ref. [4] and (ii) the actual densities obtained from the database in accordance with the modelling method adopted in Ref. [7]. The I-indices of the building materials were also calculated and compared with the absorbed gamma doses determined using the two different calculation methods. The absorbed gamma doses were compared and clearly showed that without density consideration the calculated dose is significantly higher in the case of low density building materials (see Fig. 2(a)). For densities less than 1000 kg/m^3, the overestimation can be as high as 60–70%. This is the reason why, with density consideration, the calculated I-indices correspond to a lower dose rate, which clearly proves the overestimation of I-index in connection with generated dose rate (see Fig 2(b)).

4. CONCLUSION

It was found that in most cases the application of the I-index approach without density consideration provides a significant overestimation of the dose arising from building materials. It means that the I-index provides a conservative and superficial approximation. In the case of building materials with low density, such as commonly used cellular lightweight concrete bricks, this can result in a significant overestimation of dose and an unnecessary restriction on their use.

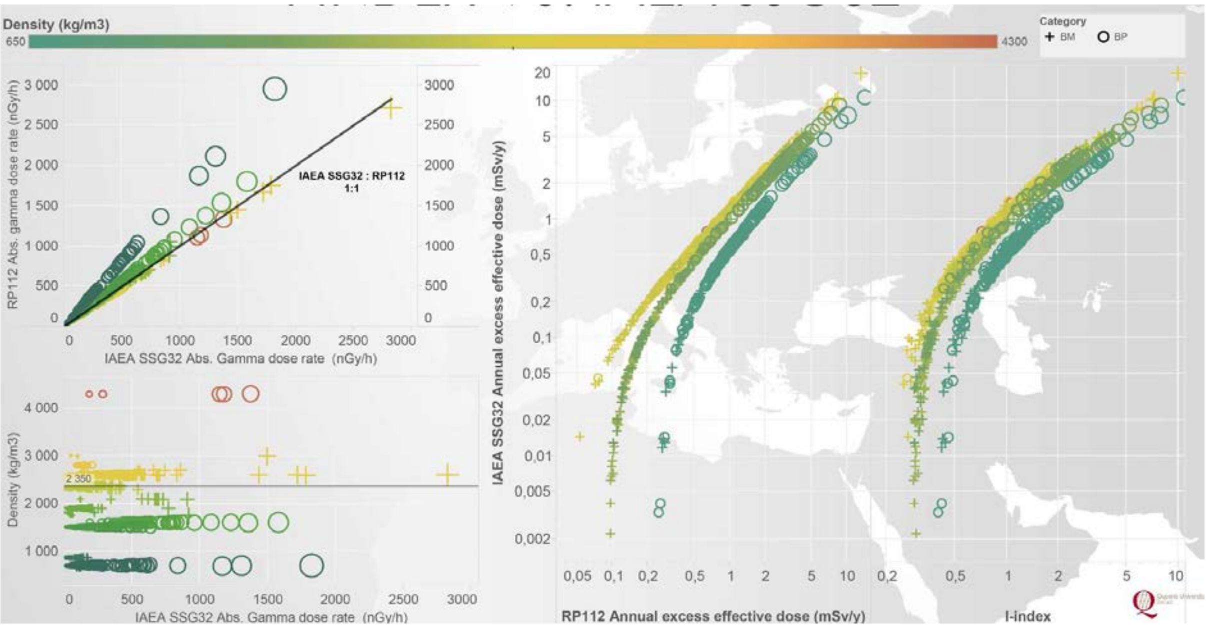

FIG. 2(a). Overestimation without density consideration of absorbed dose.

FIG. 2(b). Annual dose excess calculated with different methods in the function of I-index.

ACKNOWLEDGEMENT

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Poster Presentation

REACTIVE BARRIER DESIGN TO MITIGATE ACID MINE DRAINAGE

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Abstract

Mining and metallurgical processes generate various types of effluents that require treatment before discharge. A common environmental issue related to mining activity is the occurrence of acid mine drainage. This results from the presence of sulphide ores in a water–oxygen medium. Despite being thermodynamically favourable, this process can be accelerated by sulphate oxidizing bacteria. The Osamu Utsumi mine located in Caldas, Minas Gerais (the first uranium mine in Brazil) ceased operations in 1995 and presents this environmental problem with the acid drainage being generated in the waste rock piles. In this preliminary study, column tests were performed in the laboratory using crushed waste rock with a view to designing a permeable reactive barrier system consisting of sand and lime. The aim of this study was to assess the efficiency of reactive barriers using limestone to control acid mine drainage and thus the leaching of uranium and other metals by the acid effluent.

1. INTRODUCTION

The Osamu Utsumi mine, located in the mining–industrial complex of the Poços de Caldas Plateau in southern Minas Gerais state, was the first uranium mining and ore processing venture to operate in Brazil. The complex belongs to Brazilian Nuclear Industries (INB) [1]. INB initiated its mining activities in 1982 as an open pit mine, excavating 85 million m^3 of ore and waste rock in its 12 years of operation and attaining a daily ore production of 2500 t. The mine is now closed and undergoing decommissioning. Currently, the mine relies on a containment system for drainage collection and a treatment system for the acid effluent generated. During the production process, a significant environmental liability was created, due mainly to the generation of acidic water in waste rock piles known as 'bota-foras'. Exposure of sulphide ores to the combined action of water and oxygen, as was the case here, produces sulphuric acid. This process is accelerated by the acidophilic bacteria *Thiobacillus ferrooxidans* and *Thiobacillus sulfoxidans*. The acidic effluent then dissolves metals contained in the waste rock. The aim of this study was to evaluate the efficiency of limestone as a reactive barrier in the remediation of acid drainage in this particular situation, using samples of waste rock collected from the INB site.

2. METHODOLOGY

The samples were collected from the waste rock pile Bota-Fora 4. In the laboratory, they were crushed to a particle size of less than 4 mm and then homogenized. Percolation columns were identified as PER-00, PER-50A, PER-50B, PER-100A and PER-100B. These identification numbers refer to the amount of limestone used in each column. Thus, 50% $CaCO_3$ (1 g of limestone) was used in columns PER-50 while 100% $CaCO_3$ (2 g of limestone) was used for columns PER-100. The latter was the amount required to neutralize the H_2SO_4 that theoretically could be generated, as determined by the method described in Ref. [2]. The experiments were assembled in 4.5 cm high glass columns, as illustrated in Fig. 1.

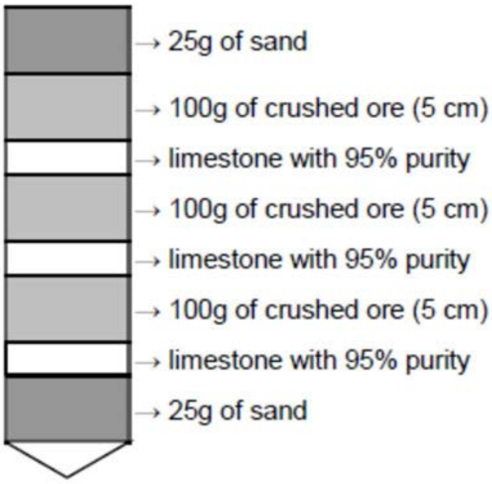

FIG. 1. Schematic composition of the columns.

The test procedure was to percolate water through the assembled columns while monitoring the chemical and physicochemical parameters of the percolated water. A water volume of 100 mL, representing 62.9 mm of rain, was added at the top of the columns. After percolation of each 100 mL aliquot of water, its pH was determined and each sample was preserved by the addition of 1 mL of HNO_3. The samples were identified and sent for chemical analysis. The use of column PER-00 was aimed at the assessment of the acid drainage generation in a Bota-Fora 4 sample which did not suffer treatment or had a limestone layer added. The water used for percolation had an average pH value of 6.3.

3. RESULTS AND DISCUSSION

3.1. Hydrogen potential (pH)

The pH values observed during the experiment are shown in Fig. 2. The average pH values observed for each column were:

PER-00:	3.32,
PER-50A:	5.99,
PER-50B:	6.18,
PER-100A:	6.82,
PER-100B:	6.84.

The pH values for columns PER-100A and PER-100B were the highest, indicating that the columns using 2 g of limestone were more efficient.

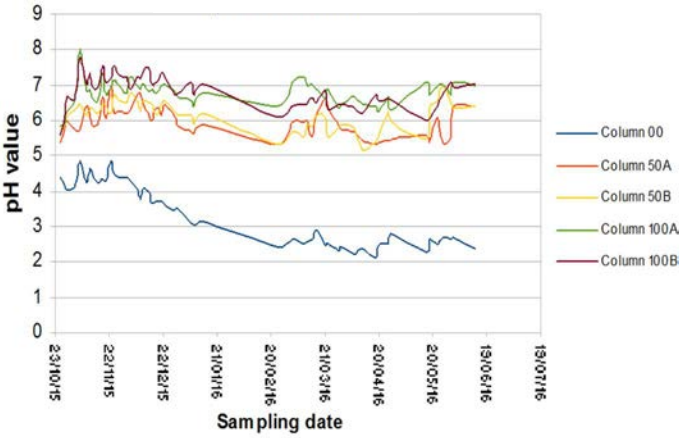

FIG. 2. Leachate pH versus time.

3.2. Sulphate

The sulphate levels observed in the leachate are shown in Fig. 3. The PER-00 column gave a higher concentration of sulphate ions (SO_4^{2-}) compared with the others, indicating the efficiency of limestone in reducing pyrite oxidation. This seems to result from the removal of O_2 by the CO_2 gas produced by the carbonate ion decomposition. The average sulphate concentrations for each column were:

PER-00:	462 mg/L,
PER-50A:	360 mg/L,
PER-50B:	340 mg/L,
PER-100A:	266 mg/L,
PER-100B:	280 mg/L.

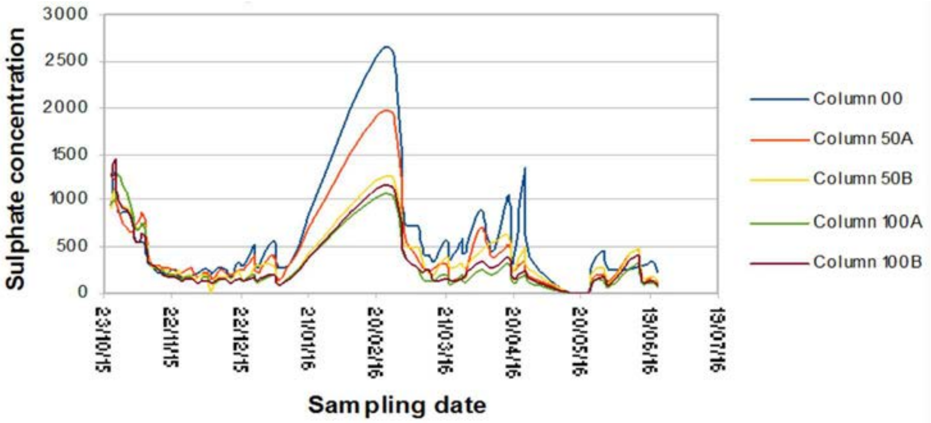

FIG. 3. Leachate sulphate concentration versus time.

3.3. U_3O_8 concentration

The U_3O_8 concentrations observed in the leachate are shown in Fig. 4. Columns PER-50 and PER-100 displayed similar behaviour, in that they were both effective in retaining U_3O_8. The average concentrations for each column were:

PER-00: 0.126 mg/L,
PER-50A: 0.008 mg/L,
PER-50B: 0.016 mg/L,
PER-100A: 0.006 mg/L,
PER-100B: 0.006 mg/L.

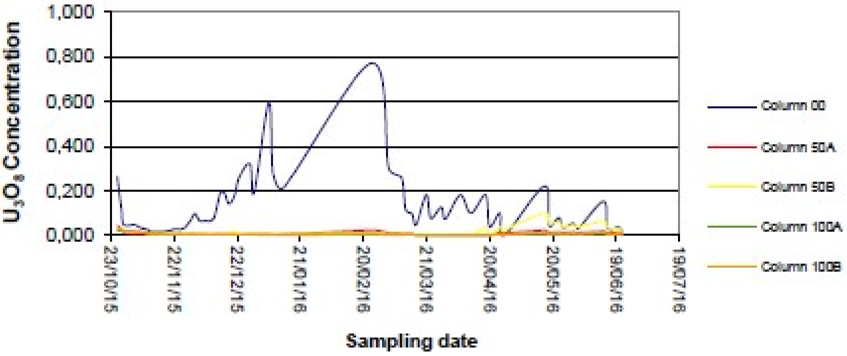

FIG. 4. Leachate U_3O_8 concentration versus time.

4. CONCLUSIONS

The results obtained in this study demonstrate the efficiency of the reactive barrier using limestone as a remediation measure against acid mine drainage. The best results were obtained in columns PER-100A and PER-100B. The pH values of the leachates in columns PER-100A and PER-100B were close to 7, while the use of limestone demonstrated efficiency in reducing pyrite oxidation. The same columns were also effective at retaining U_3O_8. This study was preliminary in nature and other experiments need to be conducted on a larger scale in order to better represent the behaviour of the waste rock piles, as well as to test different configurations of limestone application.

ACKNOWLEDGEMENTS

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Poster Presentation

FABRICATION OF TITANIUM DIOXIDE SUPPORTS FOR CERAMIC MEMBRANES USED IN THE TREATMENT OF NUCLEAR WASTE

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Abstract

In the nuclear fuel cycle, nuclear waste is generated during the production and application of radioisotopes, as well as during the processing of materials containing radionuclides of natural origin. The separation process used for this waste is a combination of two mechanisms: size exclusion and ion ionic charge. Nanofiltration has a negative electrical charge in an aqueous medium, facilitating the rejection of multivalent anions resulting from an increase in the electrostatic repulsion forces. The ceramic porous membrane used for this purpose has specific thermal and chemical stability requirements. This work investigates the effect of sintering temperature on the permeability and mechanical properties of a ceramic support produced from titanium dioxide with an approximate particle size of 21 nm, used to treat radioactive waste of medium and low activity. For this purpose, drying of the ceramic in a spray dryer was used, so that the suspension could be accomplished by an appropriate method of pressing, resulting in good product properties after sintering. The porosity of the samples was determined by means of geometric measurements and the application of nuclear techniques (gamma scattering). The results showed that the final porosity obtained was satisfactory for nuclear waste treatment.

1. INTRODUCTION

A membrane is a selective barrier between two phases, which allows the preferential transport of one phase over another, usually under a pressure gradient. Owing to the progressive increase in the cost of energy and the growth of research on separation processes, conventional techniques such as distillation, crystallization and filtration have been replaced by the use of membranes. Furthermore, the use of membranes constructed from ceramics rather than polymers enables a greater diversity of applications, owing to the ability to withstand temperatures exceeding 150°C and high corrosion resistance. Moreover, the use of substrates, better known as supports, is required, in order to compensate for the brittleness and the thin nature of the ceramic membranes, promoting improved mechanical properties. This component should also have reasonable chemical inertness, provide a convenient surface for deposition of another layer and negligible hydraulic resistance [1].

The initial stage of membrane fabrication involves the choice of a suitable ceramic powder, which determines the properties of the final product, taking into account characteristics such as: purity, particle size distribution and reactivity. For example, the particle size determines the pore diameter. In addition, the material comprising the ceramic membrane and its chemical affinity are important determinants of separation performance. To achieve high permeability associated with good selectivity, asymmetric structures are used, consisting of discrete layers with pore sizes decreasing from the base to the surface. Ceramic membranes demonstrate qualities such as high chemical resistance, performance over a wide pH range, good permeability, high thermal stability, high flow separation and filtration process associated with specific surface properties. Membrane technology becomes especially interesting with regard to the treatment of effluents because of the low energy cost, lack of chemicals, retention

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† Deceased.

of undesirable compounds and simple operating procedure. Another important application is in control and environmental monitoring, in that ceramic membranes allow the removal of precipitated radionuclides and the recycling of refrigerants used in machining [2].

Substances that decompose during the firing step are typically used, in order to facilitate the proper choice of manufacturing process and to provide for the application of a membrane format associated with the proper selection of additives for the generation of pores. The importance of this stage is due to the fact that the size and the pore volume influence the selectivity and permeability of the membrane, thereby defining the application possibilities of this material. For a given porosity value, however, the permeability may be different, since it depends on how the pores are distributed in the sample. The volume fraction of voids should be between 30 and 40% (ideally 35%) for filtration applications [2]. The submicron ceramic particles typically used are inconvenient because of poor flow properties, causing the particles to agglomerate and exhibit irregularly shaped low packaging. This results in the inability to automate the process, reducing productivity. To circumvent this problem, the particles undergo a process known as granulation, which generates spherical, homogeneous and regular shaped granules. The main method employed is drying by spray drying, in which an aqueous suspension of ceramic powder is sprayed into a heated chamber, giving rise to rapid water evaporation and granule formation. The process also allows for the homogenous distribution of the additives responsible for improving certain properties of the final product, such as mechanical strength. The variables involved in the spray drying process are moisture suspension, degree of deflocculation, spraying characteristics, drying temperature and organic additives. These factors affect characteristics such as the size distribution of the granules, flow properties, degree of deformation, density and mechanical strength [3].

Additives are added before the drying of this suspension by spray dryer and classified according to their purpose, i.e. dispersants, binders, pore formers and surfactants. It is essential that they are organic in nature, so that decomposition is possible below the sintering temperature to avoid contamination of the product. Lubricants are important in the forming step and in the workpiece extraction step. Polymers of hygroscopic nature are usually used to confer mechanical strength on the compressed parts to be handled without fracturing [2]. Pressing is the most commonly used process for obtaining ceramic parts as it facilitates high productivity. It is important to understand variables such as relative humidity and temperature because they affect the incidence of defects in the product. Pressing involves the application of pressure in a granular material confined within a rigid matrix. It can be divided into three stages: (i) filling the matrix, (ii) powder compaction, and (iii) extraction of the piece. One of the methods most commonly used in this case is the uniaxial method, in which the compaction is performed in a matrix by the application of load by a single shaft through a hole. The load may be unidirectional (only one punch in operation) or bidirectional (two punches operational simultaneously), suitable for most applications since it minimizes the pressure gradient and therefore reduces density variations along the workpiece [2]. Sintering is the final step in the process and consists of heating and particle consolidation in the form of compacted powder, during which the size and shape of pores change as a function of temperature and time [2].

2. MATERIALS AND METHODS

Titanium dioxide (150g in the rutile phase of a commercial product called R-KB Tiona 2 BR) was added gradually to 1 L of distilled water. Flocculation of the ceramic powder was avoided by stirring with a rotor at 510 rpm. Then, 7.5 g of a pore forming agent in the form of potato starch (Yoki) was added. The preparation process is shown in Fig. 1. This was followed by the addition of 20 mL of a 10% solution of PVA binder (prepared using a magnetic stirrer at 80°C). Then, to prevent agglomeration of the titanium dioxide particles, LA dispersant was added, taking care not to add more than 0.5 g in order to avoid an undue increase in the viscosity of the suspension. Next, 0.5 L of distilled water was added to obtain a better viscosity and, finally, one drop of Lambeti SE 47 antifoam agent was added to ensure physical stability.

To obtain the correct granule size distribution and complete drying of the solution, the prepared suspension was poured into an APV Anhydro AS spray dryer. The drying operation was carried out at a feed rate of 35 mL/min, an inlet temperature of 149.8°C and an outlet temperature of 108°C. The machine had two granule outlets, one for coarse granules and the other for finer granules. The drying process was observed to be efficient since no pulp remained attached to the inner walls of the equipment and there was no moisture in the product.

FIG. 1. Preparation of the titanium dioxide suspension.

FIG. 2. The spray dryer (photograph obtained from product manual).

Uniaxial and unidirectional pressing was then performed at a pressure of 1.5 N/cm^2, yielding product in the form of 15 'tablets', each made from 1.5 g of the spray dried ceramic powder. Sintering was carried out in an oven (see Fig. 3) at three temperatures: 1050°C, 1100°C and1150°C (five elements at each temperature). Finally, the tablets were measured and weighed in order to calculate the density and porosity from Eqs (1) and (2) [4].

$$\text{Theoretical density} = \frac{\text{Real density}}{\text{Manufacturer density}} \times 100 \quad (\%) \tag{1}$$

$$\text{Porosity} = 100 - \text{Theoretical density} \quad (2)$$

FIG. 3. The sintering oven.

In order to determine the porosity by gamma spectroscopy, it was first necessary to determine the attenuation coefficient of the material free from pores, using Eq. (3) [5].

$$\mu_a = \frac{1}{d} \cdot \ln \frac{I_0}{I} \quad (3)$$

where μ_a is the coefficient of attenuation of the material, d is the thickness of the material, I_0 is the intensity of the beam array incident on the material and I is the intensity of the beam transmitted through the material. Thus, the total porosity is calculated from Eq. (4) [5]:

$$P_t = \frac{\mu_a - \mu_b}{\mu_a} \times 100 \quad (\%) \quad (4)$$

where μ_b is the coefficient of attenuation of the porous material submitted to the gamma radiation beam. The assembly of the data acquisition system is illustrated in Fig. 4.

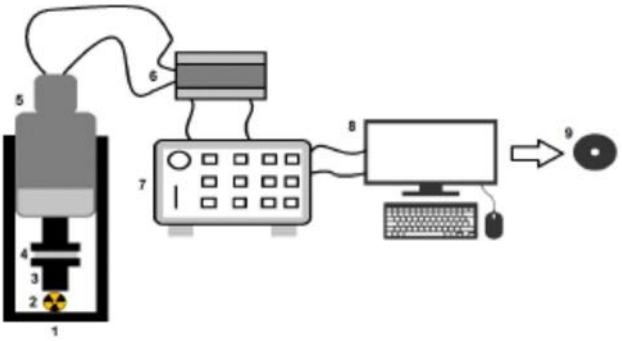

FIG. 4. Gamma radiation transmission system: (1) Lead shield, (2) radioactive source, (3) collimator, (4) sample, (5) scintillator detector, (6) preamplifier, (7) multichannel analyser, (8) computer for data acquisition via software, (9) removable media data storage.

3. RESULTS AND DISCUSSION

From the measurements of the relevant physical characteristics, the density and porosity of each sample were calculated. The results are shown in Table 1 and Fig. 5. The value of the manufacturer density was 4.1 kg/m^3. A brief review of Table 1 shows that, with increasing sintering temperature, the density decreases and the porosity increases. This is in perfect agreement with theory — since the sintering process is thermally activated, the higher the temperature, the greater the energy supplied to the ceramic material and therefore the greater the degree of sintering, resulting in a higher density. Moreover, tablets sintered at the same temperature exhibit similar porosity, demonstrating that the composition of the granules was homogeneous and therefore that the ceramic processing was adequate.

TABLE 1. DENSITY AND POROSITY CALCULATED FROM PHYSICAL MEASUREMENTS

Sample	Mass (g)	Diameter (mm)	Thickness (mm)	Density (kg/m^3)	Theoretical density (%)	Porosity (%)	Sintering temperature (°C)
1	1.37	23.81	1.52	2.02	49.27	50.73	1050
2	1.38	23.86	1.50	2.06	50.19	49.81	1050
3	1.38	23.83	1.53	2.02	49.39	50.61	1050
4	1.38	23.78	1.54	2.03	49.49	50.51	1050
5	1.41	23.79	1.56	2.04	49.77	50.23	1050
6	1.38	22.87	1.44	2.34	57.09	42.91	1100
7	1.40	22.86	1.48	2.29	55.92	44.08	1100
8	1.39	22.85	1.46	2.32	56.47	43.53	1100
9	1.38	22.82	1.46	2.32	56.53	43.47	1100
10	1.38	22.84	1.46	2.31	56.44	43.56	1100
11	1.39	22.04	1.42	2.58	62.92	37.08	1150
12	1.39	22.08	1.42	2.56	62.37	37.63	1150
13	1.38	22.07	1.40	2.58	62.99	37.01	1150
14	1.38	22.01	1.39	2.61	63.55	36.45	1150
15	1.39	22.02	1.41	2.58	63.00	37.00	1150

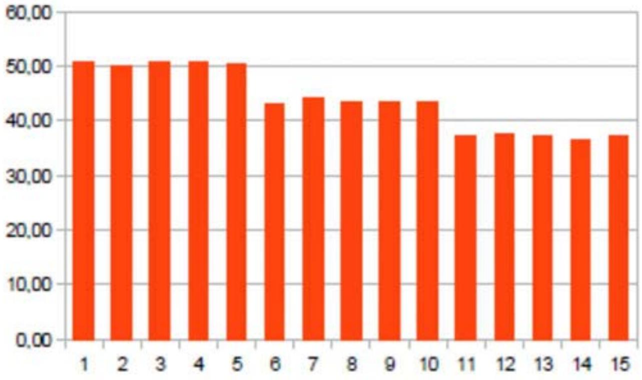

FIG. 5. Porosities (%) of the 15 samples.

The gamma spectroscopic determination of porosity yielded a value 57.134% (uncertainty 7.946%), which compared well with a value of 50.006% (uncertainty 0.014%) determined from physical measurements. The two methods are therefore compatible. However, the experimental uncertainty is greater for the spectroscopic method. This is because of the reduced number of regions used for the calculation of the porosity (three only). In addition, the porosity values calculated from physical measurements may be underestimated because in practice the solid is not perfectly homogeneous, leading to a less accurate characterization of the bulk material.

4. CONCLUSIONS

Since all samples had the same concentration of pore forming agent, it is clear that this is not the only parameter governing porosity — it obviously depends also on the sintering temperature, which influences the consolidation of the material and thus its void fraction. It is concluded that gamma spectroscopy as a technique for determining porosity is worthy of consideration since it can cover a greater number of regions of the test sample, it is non-destructive, and it is sensitive to dimensional variations of the sample material.

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Poster Presentation

URANIUM RECOVERY FROM ACID MINE DRAINAGE TREATMENT RESIDUE

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Abstract

The generation of acid mine drainage from mining areas due to the presence of sulphide minerals (usually pyrite, FeS_2) in contact with oxygen and water, is a huge environmental problem. The Osamu Utsumi Mine located in Caldas, Minas Gerais, the first uranium mine in Brazil and which ceased operations in 1995, presents this environmental issue. The acid solution is produced from the waste rock piles and leaches residual metals, including uranium. This effluent is being treated continuously with lime, the residue from which, an alkaline mud, is deposited into the mine pit. This alkaline mud contains uranium and rare earths and several projects are being carried out in order to extract these products. This paper describes the efforts to develop an alkaline leaching process to extract and concentrate uranium liquor from this residual material. As its U_3O_8 concentration ranges from 1800–3000 mg/L, this recovery is interesting not only from a commercial point of view but also from the environmental one, as it enables the material to be disposed of in a proper way during the mine closure process.

1. INTRODUCTION

In 1952, Brazil began its first exploration in search of uranium and thorium ores. About a third of the country was explored, which led to the identification of the country's uranium reserves (309 000 t of U_3O_8) as being the seventh largest in the world [1]. The first uranium mine in Brazil was the Osamu Utsumi Mine, located on the Poços de Caldas Plateau in the city of Caldas, Minas Gerais State and operated by the state owned company Brazilian Nuclear Industries (INB). At the site, INB produced uranium concentrate in the form of ammonium diuranate, this being the first step in the nuclear fuel cycle. Mining operations ceased in 1995, but the environmental problem resulting from the generation of acidic drainage within the mine pit and two waste rock piles known as Bota-Foras 4 and 8 still occurs at the site. The critical situation faced by Caldas is not unique, with this problem occurring at other mines, especially those with ores containing high sulphide concentrations. The high sulphide concentration along with the presence of water and oxygen generates sulphuric acid, which may lead to the dissolution of metals such as uranium, rare earths and iron [2]. The acid water currently generated on the site has been treated by the addition of hydrated lime, producing an alkaline residue consisting of a matrix of calcium sulphate. As this residue contains commercially viable concentrations of uranium, the aim of this study was to recover the uranium in order to reduce the associated environmental problem as well as to provide a safer option for disposal of the residual material as waste.

2. METHODOLOGY

Samples of residue were collected from INB at Caldas and then dried, crushed and homogenized before leaching. The characterization of these samples in the laboratory has been reported elsewhere [3], with the following results:

U_3O_8	0.318 ± 0.016,
Al	4.600 ± 0.500,
Ca	15.200 ± 1.900,
Ce	0.947 ± 0.061,
Dy	0.011 ± 0.001,
Eu	0.007 ± 0.001,
Fe	0.728 ± 0.060,
Gd	0.011 ± 0.001,
La	1.340 ± 0.100,
Mn	2.300 ± 0.120,
Nd	0.423 ± 0.001,
Pr	0.250 ± 0.014,
Sm	0.028 ± 0.001,
Y	0.131 ± 0.013,
Yb	0.003 ± 0.001,
Solids	17.710 ± 0.050,
TR (totals)	3.150 ± 0.014.

In the present study, factors such as solids percentage, temperature, reaction time period and type of leaching agent were tested and evaluated in order to determine the best operational conditions for leaching. For all leaching tests it was decided to maintain the solids ratio at 10%, which, according to Refs [3, 4], is the optimum value for this operational parameter. For all tests, beakers of 400 mL under mechanical agitation were utilized. The agitator employed was a SCILOGEX OS40-Pro with metal propellers and a SCILOGEX MS7-H550-S heating plate. After each test the solution was filtered to obtain the uranium leachate, which was subsequently sent for chemical analysis.

3. RESULTS AND DISCUSSION

3.1. Leaching agent

The leaching agent is the chemical compound responsible for effectively releasing the uranium found in the residue, in order to allow it to be concentrated later on. Because it is an alkaline leaching process, the agent to be used must have the characteristics of a base. Leaching tests were conducted for 4 h, initially at 70°C, to identify which agent or which mixture of agents provided the best results for uranium dissolution. The agents tested were:

- Sodium carbonate 1M,
- Sodium bicarbonate 1M,
- Sodium carbonate 1M + sodium bicarbonate 1M (1:1 proportion),
- Sodium carbonate 1M + sodium bicarbonate 1M (1:3 proportion).

The results obtained are shown in Fig. 1. The 1:3 mixture of sodium carbonate and sodium bicarbonate gave the highest uranium recovery (>85%).

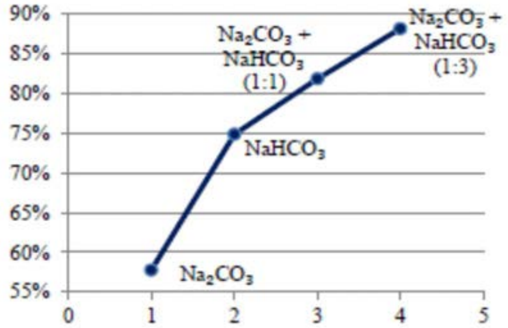

FIG. 1. Dissolution of U_3O_8 for various leaching agents.

3.2. Leaching temperature

Leaching tests were carried out at temperatures of 25°C (room temperature), 50°C, 70°C and 90°C, maintaining the same leaching time of 4 h and using the 1:3 mixture of sodium carbonate and sodium bicarbonate as the leaching agent. The results are shown in Fig. 2. It is noteworthy that, although higher dissolution is observed at 90ºC, greater evaporation also occurs, resulting in the need for more intensive volume control.

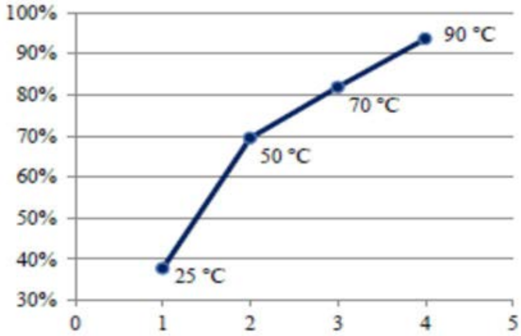

FIG. 2. Dissolution of U_3O_8 for various leaching temperatures.

3.3. Leaching time

The identification of the best time period for uranium dissolution is an important aspect of this study, for it involves energetic output, process feasibility and other relevant aspects. For this parameter, experiments were carried out for reaction times of 1, 2, 4 and 6 h, keeping the temperature at 70°C and using the 1:3 mixture of sodium carbonate and sodium bicarbonate as the leaching agent. The results are shown in Fig. 3. A point to be emphasized is the lower dissolution after 6 h compared with that after 4 h. Such behaviour may be due to a change in the relationship between carbonate and bicarbonate, which may occur with increasing reaction time, leading to lower uranium dissolution.

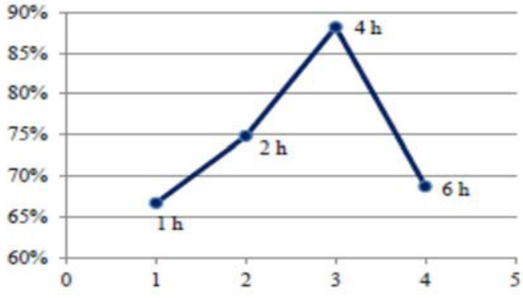

FIG. 3. Dissolution of U_3O_8 for various leaching times.

Once the optimum values for all parameters were determined, a leaching test was conducted, combining all the best parameters previously identified. This led to an 88% uranium recovery from the residue.

4. CONCLUSIONS

The results show that the ideal conditions for uranium recovery from the residue by means of alkaline leaching are:

- An aqueous solution of sodium carbonate and sodium bicarbonate at a ratio of 1:3,
- A reaction time of 4 h,
- A reaction temperature of 70ºC,
- A solids percentage of 10%.

Under these conditions, 88% recovery of uranium was achieved. It is planned that the current study will continue with the aim of increasing the final uranium concentration in the leachate to a value of 1 g/L through leaching in a recycling mode. Another point to be investigated is how to decrease the amount of carbonate in relation to uranium, as the literature indicates a lower uranium extraction for a high ratio between these components.

ACKNOWLEDGEMENTS

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NATURAL RADIOACTIVITY

(Session 4)

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Invited Paper

UNSCEAR REVIEW OF RADIATION EXPOSURE DUE TO ACTIVITIES INVOLVING NORM

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Abstract

Industries and other human activities that process NORM give rise to occupational and public exposures. To help estimate the average and collective annual effective doses due to these activities, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) published in 2008 the results of literature reviews and expert group reports on the sources and effects of radiation, including NORM sources. It turns out that for occupationally exposed workers the average annual dose in NORM industries and activities is around 3 mSv and the annual collective dose is around 40 000 man Sv. The information on public exposure from these activities is sparser, but maximum annual doses can exceed 1 mSv.

1. INTRODUCTION

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has periodically compiled data and trends on occupational and public exposure due to industrial and other activities involving NORM. This paper summarizes the information available in UNSCEAR up to the present date. The information is based on the 2008 UNSCEAR report Sources and Effects of Ionizing Radiation [1]. UNSCEAR envisages compiling exposure information in due course for an update to the 2008 sources report after completing a highly pertinent re-evaluation of the risks of lung cancer due to exposure to radon. The IAEA also has a number of publications available on radiation protection and NORM [2]

2. OCCUPATIONAL EXPOSURE

The data up to 2002 summarized in Fig. 1 shows clearly that, although an occupationally exposed worker in an industry or activity exposed to natural radiation receives an average annual effective dose of around 3 mSv, owing to the large number of workers so exposed the annual collective dose for these workers is considerably higher than that from the sum of all other occupationally exposed workers. The majority of this annual collective dose comes from mining activities (other than uranium mining and ore processing), as can be seen from Table 1.

The number of mine workers occupationally exposed depends on the economic activity in this area and it is expected that this annual collective dose will increase as new information on occupational exposure becomes available from mining operations where individual monitoring had not previously been carried out and effective dose estimation was not available. Other major sources of occupational exposure to NORM are found in the oil and gas sector, phosphate fertilizer production, zircon milling, rare earth and titanium processing industries. Apart from mining and ore processing, other activities that result in occupational NORM exposure are shown in Table 2.

The factors that affect uncertainties in NORM exposure data are the reliability of individual monitoring methods and data recording, evaluation of anomalies, such as unexpectedly high or low values, subtraction of background doses, the choice as to who should be monitored and whether or not internal exposures are included in the effective dose estimate.

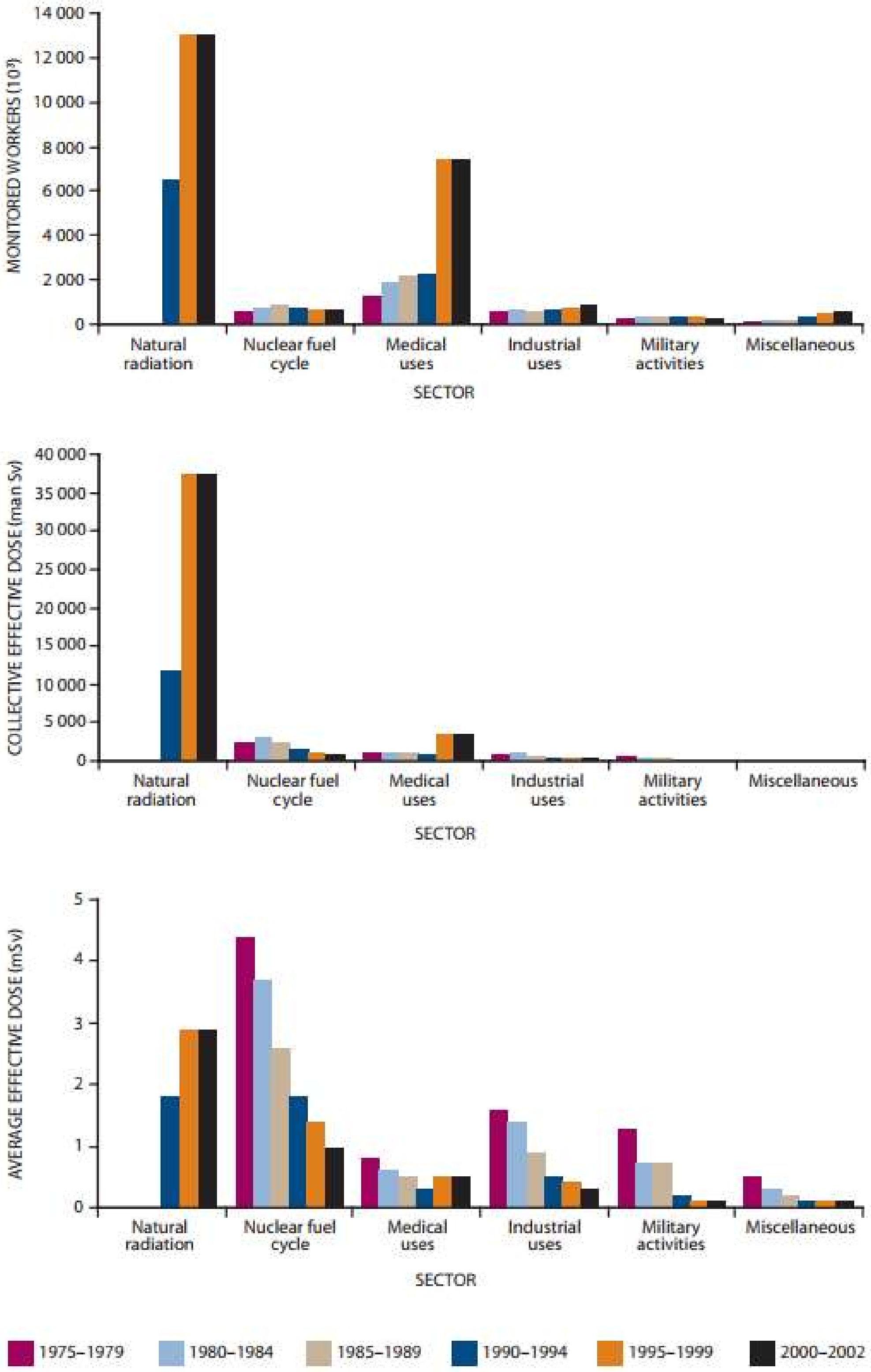

FIG. 1. Summary of number of monitored persons and the annual collective and average effective doses to occupationally exposed workers from 1975 to 2002.

TABLE 1. ANNUAL DOSES RECEIVED BY COAL MINE WORKERS IN CHINA

Type of coal mine	Average annual effective dose (mSv)	Collective dose (man Sv)
Large sized	0.28	280
Medium sized	0.55	550
Small-sized	3.3	13 200
Bone coal	10.9	545
Average	2.4	14 600

TABLE 2. OCCUPATIONAL RADIATION PROTECTION DATA FROM GERMANY FOR RADON INHALATION IN WORKPLACES OTHER THAN MINES

Workplace	Period	No. of workers		Annual collective effective dose (man Sv)	Average annual effective dose (mSv)	
		Monitored	Measurably exposed		Monitored workers	Measurably exposed workers
Spas	1995–1999	2	2	0.01	4.77	4.77
	2000–2002	4	2	0.01	4.09	4.47
Waterworks	1995–1999	128	75	0.24	1.85	3.12
	2000–2002	81	47	0.11	1.39	2.50
Tourist caves and visitor mines	1995–1999	135	101	0.31	2.26	3.01
	2000–2002	135	87	0.23	1.76	2.63

3. PUBLIC EXPOSURE TO NORM

The public is exposed to NORM through uranium mining and ore processing, non-uranium metal mining and smelting, the phosphate industry, coal mines and power generation from coal, oil and gas extraction, rare earth and titanium oxide industries, zirconium and ceramic industries, applications using natural radionuclides (typically radium and thorium) and the disposal of building material. For public exposure the data are very sparse and site-specific. The main exposure pathways are through the inhalation of dust and radon, contamination of groundwater with radium isotopes and external exposure to slag with high thorium content.

For uranium mining and ore processing, the most relevant sources of exposure are (i) radon and its progeny released from open pits and underground mines, (ii) in situ leaching operations and (iii) process tailings. The release of ^{226}Ra to liquid pathways is also relevant and careful consideration must be given to the use of contaminated land for building. For the phosphate industry, the processing of phosphate rock may generate emissions of ^{238}U and ^{226}Ra. Phosphogypsum stacks, the use of phosphate fertilizer and the use of phosphogypsum in building materials are also possible sources of exposure to radon. Table 3 shows estimated doses to the public due to industrial releases of NORM in the United Kingdom.

TABLE 3. DOSES TO THE PUBLIC DUE TO INDUSTRIAL RELEASE OF NORM IN THE UNITED KINGDOM

Industry	Discharge route	Pathway	Annual dose (μSv)	
			Critical group	General public
Coal-fired power station	Atmospheric release via stack	All	1.5	0.1
	Building material made from ash	Radon inhalation External	600 900	
Oil and gas extraction	Authorized discharges to sea, and scales	Ingestion of seafood and external exposure due to fishing gear	<30	
Gas-fired power station	Atmospheric releases via stack	All	0.75	0.032
Steel production	Atmospheric releases via stack	All	<100	<2
	Building material made from slag	Radon inhalation External	550 800	
Zircon sands	Atmospheric releases via stack	Inhalation	<1	<1

4. SUMMARY OF OCCUPATIONAL AND PUBLIC EXPOSURES

For occupational exposures the collective dose is around 40 000 man Sv annually (2008), with around 17 000 man Sv from coal mining, 14 000 man Sv from other mining and 6 000 man Sv from radon in other workplaces. The average individual dose in these activities is about 3 mSv (2008). There are wide variations depending on local circumstances; some mines give rise to annual effective doses of tens of millisieverts, depending on the type of mine, geology and working conditions.

Public exposure is normally low, but considerable numbers of people can be exposed. Doses of up to a few millisieverts annually can be measured for a few scenarios, e.g. use of sludge from water treatment as fertilizer, use of waste products for building material. There is at present no consistent approach to make a reliable global assessment of inventories and exposures. There is a diversity of ores with low levels of radionuclides from the uranium and thorium chains concentrated in products, by-products and waste and conventional mining that leads to huge volumes of material with enhanced concentrations of radionuclides of natural origin, making a challenge for disposal operations and site restoration.

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Poster Presentation

NATURAL RADIOACTIVITY IN SEDIMENTS AND WATER OF CLOTILDE BRIOZZO LAGOON

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Abstract

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Clotilde Briozzo lagoon coastal sediments was determined to evaluate the external hazard index, the outdoor annual effective dose and the radium equivalent activity in order to determine if those represent a risk for inhabitants. Each sediment sample was dried and placed in a 500 mL Marinelli beaker. After approximately four weeks in order to reach secular equilibrium of ^{238}U and ^{232}Th series, samples were measured by gamma spectrometry with a high purity germanium detector. IAEA reference materials were used for efficiency calibration. The ^{238}U series was studied by the photopeaks of ^{214}Bi (609.3 keV) and ^{214}Pb (295.2 keV). The ^{232}Th series was evaluated by the photopeak of ^{228}Ac (911.1 keV) and ^{40}K was evaluated by its own photopeak of 1460.8 keV. The ranges of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found to be 14.92–67.86, 41.91–131.20, and 88.90–168.08 Bq/kg, respectively. The outdoor annual effective dose equivalent, the radium equivalent activity and the external hazard index associated with the samples were evaluated. Furthermore, the gross alpha and gross beta activities in water were also determined using a low background gas proportional counter. The average activity was 0.060 Bq/L.

1. INTRODUCTION

Clotilde Briozzo Lagoon is a coastal lagoon located in Rocha state, Uruguay. It is strategically located from the point of view of research, because it is located on black sand ore that has not yet been disturbed by human activity. This ore was studied in the 1960s [1] and the 2000s [2, 3] by government institutions. These studies reported on the heavy mineral content of the sand — 50% ilmenite, 20% magnetite, 5% zircon, 1% rutile and 0.6% monazite. The monazite in turn contains 4.75% ThO_2 and 0.18% UO_2. However, the studies did not determine the concentrations of radionuclides of natural origin and did not evaluate the radiological significance of these deposits, bearing in mind that radionuclides in the ^{238}U and ^{232}Th series and ^{40}K, together with those formed by the interaction of cosmic rays in the Earth´s atmosphere, represent the majority of radionuclides of natural origin [4] and are responsible for 85% of the exposure received by the world population [5].

There are recent reports of gross alpha and gross beta activity concentrations in sand and water in Barra de Valizas [6] and the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the sand beaches of Rocha [7], all places forming part of the Uruguayan coast. These reports included activity measurements in Barra de Valizas and Aguas Dulces which are situated near the Clotilde Briozzo Lagoon and also are located on black sand ore. These previous reports suggest that it is necessary to know the activity concentrations of these radionuclides all along the coast and especially in Clotilde Briozzo Lagoon — this was the aim of this work.

2. METHODS

In order to evaluate the levels of natural radioactivity, sediment and water samples were collected from three locations on the Clotilde Briozzo Lagoon: (i) 34°17'35.87"S, 53°48'10.45"W, (ii) 34°17'35.1"S, 53°48'11.16"W, (iii) 34°17'35.60"S, 53°48'11.30"W).

2.1. Sediment samples

Sediment samples were obtained at each location at a typical depth of 0 to 20 cm from the top surface layer to produce approximately 2 kg wet weight per sample. After removing stones and vegetable matter, each sample was dried at a temperature of 60°C until constant weight was reached, and passed through a standard 2 mm mesh size sieve. The homogenized samples were filled into 500 mL Marinelli beakers, hermetically sealed and stored for at least four weeks in order to attain radioactive secular equilibrium between ^{232}Th and ^{226}Ra and their short-lived progeny.

The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in the sediment samples were measured using a high resolution gamma-ray spectrometry system with a high purity germanium detector AMETEK/ORTEC GMX35P4-76-RB, having 35% efficiency and 1.75% photopeak resolution for ^{60}Co. IAEA reference materials RGU-1, RGTh-1 and RGK-1 were used for the efficiency calibration. Each sediment sample was measured for 150 000 s, with a dead time of 0.12–0.38 %. The ^{238}U series was studied by the weighted mean of the photopeaks of ^{214}Bi (609.3 keV) and ^{214}Pb (295.2 keV). The ^{232}Th series was evaluated by the photopeak of ^{228}Ac (911.1 keV), while ^{40}K was evaluated by its own photopeak 1460.8 keV. The absorbed dose rate, the annual effective dose, the radium equivalent activity and the external hazard index were evaluated using dose coefficients published in the UNSCEAR 2008 report [5].

2.2. Water samples

Water samples were taken in duplicate approximately 8–10 m away from the lagoon edge to produce approximately 6 L. For each water sample, pH was determined and nitric acid was added, then the sample was cooled to 4°C for conservation [8].

In order to determine the gross alpha and gross beta activities in the water samples, 1 L of each sample was evaporated to dryness in counting pans with the appropriate geometry for the counting system. Measurements were performed in a gas proportional Canberra LB4200 multi-detector low background alpha–beta counting system. Each sample was measured ten times with a detector live time of 600 s. Gross alpha and gross beta activity calibrations were performed with ^{241}Am (581 Bq) and ^{36}Cl (574 Bq) Eckert & Ziegler sources, respectively. Background was measured by evaporating Type II (ASTM II) water from a ThermoSmart2Pure ultra purifier under the same conditions as those for the samples and measuring the counting pans 15 times with a detector live time of 2700 s.

3. RESULTS AND DISCUSSION

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the sediment samples are shown in Table 1. The maximum value for ^{226}Ra (67.86 ± 2.08 Bq/kg) is higher than the upper bound of the worldwide range reported by UNSCEAR [2], but the average concentration is comparable with the worldwide mean. For ^{232}Th, the maximum value is 131.20 ± 10.21 Bq/kg and the average concentration is twice the worldwide mean reported by UNSCEAR but comparable with the activity concentrations for Aguas Dulces and Barra de Valizas reported elsewhere [7]. It is also comparable with other activity concentrations reported for some Brazilian beaches where similar ores have been studied [9–10].

In order to assess the radiological significance for humans, the parameters shown in Table 2 were determined. The estimated absorbed dose rate based on the sediment measurements ranges from 40.11 to 114.30 nGy/h. The mean value is higher than the worldwide mean reported by UNSCEAR. The effective dose associated with these sediments ranges from 49.22 to 140.18 μSv/a, with a mean of 80.29 μSv/a, which is higher than the worldwide mean effective dose of

70 µSv/a but comparable with the values reported for Barra de Valizas in Uruguay [7], Thailand [11], India [12] and Italy [13]. The values for radium equivalent activity and external radiation index are below the limits recommended by the OECD [14], so the use of these sediments as raw materials for building would not constitute a radiological hazard to members of the public.

TABLE1. RADIOACTIVITY IN THE SEDIMENT SAMPLES

	Activity concentration (Bq/kg)		
	Ra-226	Th-232	K-40
Sample location 1	67.86 ± 2.08	131.20 ± 10.21	88.90 ± 2.64
Sample location 2	14.92 ± 0.65	43.60 ± 3.70	165.63 ± 4.23
Sample location 3	20.91 ± 0.83	41.91 ± 3.57	168.08 ± 4.28
Worldwide range	17–60	11–64	140–850
Worldwide mean	35	30	400

TABLE 2. RADIOLOGICAL SIGNIFICANCE OF THE SEDIMENT SAMPLES

	Absorbed dose rate (nGy/h)	Effective dose (µSv/a)	Radium equivalent (Bq/kg)	External hazard index
Sample location 1	114.3	140.18	262.32	0.71
Sample location 2	40.11	49.22	90.02	0.24
Sample location 3	41.98	51.49	93.78	0.25
Worldwide mean	57	70	<370	<1

The gross alpha and gross beta activities measured in the water samples are shown in Table 3. All values are below the minimum detectable activity for alpha and below the limit for drinking water recommended by the World Health Organization for beta [15]. All water samples have a pH in the range 7.5–7.6. Although these results indicate that drinking this water would not constitute a radiological hazard in terms of alpha and beta activity, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the water and in other environmental compartments would need to be measured in order to determine transfer factors and the radiological significance for the local ecosystem.

TABLE 3. RADIOACTIVITY IN THE WATER SAMPLES

	Activity concentration (Bq/L)	
	Gross alpha	Gross beta
Sample location 1	<MDA	0.064 ± 0.017
Sample location 2	<MDA	0.061 ± 0.016
Sample location 3	<MDA	0.054 ± 0.016

Note: MDA – minimum detectable activity.

4. CONCLUSIONS

In the sediment samples, the average ^{232}Th activity concentration is higher than the worldwide mean reported by UNSCEAR, while the average activity concentration for ^{226}Ra is comparable with the worldwide mean. This agrees with the ore composition reported previously. The mean effective dose associated with the sediments is higher than the worldwide mean. The radium equivalent activity and the external index are below the recommended limits, so the use of these sediments as raw materials for building would not constitute a radiological hazard. Preliminary results of gross alpha and beta activities in the water samples indicate that drinking this water would not constitute a radiological hazard. Future work will be focused on determining transfer factors for ^{226}Ra, ^{232}Th and ^{40}K from sediments to water and other environmental compartments.

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Poster Presentation

GROSS ALPHA AND GROSS BETA RADIOACTIVITY DETERMINATION IN THE COASTAL GROUNDWATER OF ROCHA.

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Abstract

Rocha State has one of the biggest deposits of black sand in Uruguay, with approximately 7 million t of heavy minerals containing uranium, thorium and their decay progeny. Also, the region has a very rich biodiversity, with a wide variety of flora and more than 300 bird and mammal species located in different habitats and aquatic ecosystems. Groundwater samples were taken in 3 different resorts on the Atlantic coast of Rocha, where the local population exceeds 100 — in summer the population grows to three to four thousand people. Nine locations were sampled in duplicate. Every sample was evaporated to dryness and the residue was measured ten times with a Canberra LB4200 system with a detector live time of 10 min. As expected, the major beta activity was measured in Barra de Valizas, because this resort is located above a black sand deposit. All results were compared with Uruguayan standards and with the World Health Organization drinking water guidelines. The gross alpha and gross beta activities of the water samples were below the relevant limits for drinking water.

1. INTRODUCTION

Rocha State has one of the biggest deposits of black sand in Uruguay, with reserves of approximately 7 million t of heavy minerals containing uranium, thorium and their decay progeny [1–4]. Also, the region has a very rich biodiversity, with a wide variety of flora and more than 300 bird and mammal species located in different habitats and aquatic ecosystems, including a large number of fish species, whales and sea wolves in the marine ecosystems [5]. Very few measurements of radioactivity in water have been made in Uruguay [6]. The work reported in this paper is part of a groundwater radiological screening programme along the Atlantic coast of Rocha, especially a groundwater source used as drinking water by the local population and summer tourists. The presence of radionuclides in drinking water may pose significant hazards because they are deposited in the human body and gradually expose living tissues [7]. Gamma radiation has the highest penetrating power, while the effects of alpha and beta particles within the body are not very detrimental because of their relatively low ionizing potential. Gross alpha and gross beta activity determination is a screening method for water, including drinking water, as a preliminary procedure to determine whether further analysis is necessary. These measurements can be used for identifying spatial or temporal trends in the radionuclide content of water [8]. The method is applicable to the measurement of alpha emitters having energies above 3.9 MeV and beta emitters having maximum energies above 0.1 MeV [9].

2. EXPERIMENTAL

Samples were taken in duplicate at three different resorts along the Atlantic coast of Rocha. Nine locations were sampled, three in Santa Isabel de la Pedrera, four in Barra de Valizas and two in Aguas Dulces (see Table 1 and Fig.1). Every sample was evaporated to dryness in counting pans with the appropriate geometry for the counting system. Gross alpha and gross beta measurements were performed in a gas proportional Canberra LB4200 multi-detector low background alpha–beta counting system. Each sample was measured ten times with a detector live time of 10 min. Gross alpha and gross beta activity calibrations were performed with 581 Bq ^{241}Am and 574 Bq ^{36}Cl Eckert & Ziegler sources, respectively. Background was determined by evaporating Type II (ASTM II) water from a ThermoSmart2Pure ultra purifier under the same conditions as the samples and measuring the counting pans 15 times with a detector live time of 45 min.

.TABLE 1. COORDINATES OF THE SAMPLING POINTS

Sample code	Location	Coordinates
HAD02-220116	Aguas Dulces	34°16'53.29"S; 53°48'16.28"W
HAD26-161115	Aguas Dulces	34°16'26.61"S; 53°47'20.67"W
HSI01-281115	Santa Isabel de la Pedrera	34°34'1.5"S; 54°6'39.8"W
HSI05-021215	Santa Isabel de la Pedrera	34°34'7.5"S; 54°6'31.5"W
HSI07-021215	Santa Isabel de la Pedrera	34°34'13.1"S; 54°6'39.8"W
HVa01-250116	Barra de Valizas	34°20'26.3"S; 53°47'26.9"W
HVa23-161115	Barra de Valizas	34°20'16.58"S; 53°47'35.03"W
HVa24-161115	Barra de Valizas	34°20'11.92"S; 53°47'52.16"W
HVa29-161115	Barra de Valizas	34°19'54.83"S; 53°48'25.04"W

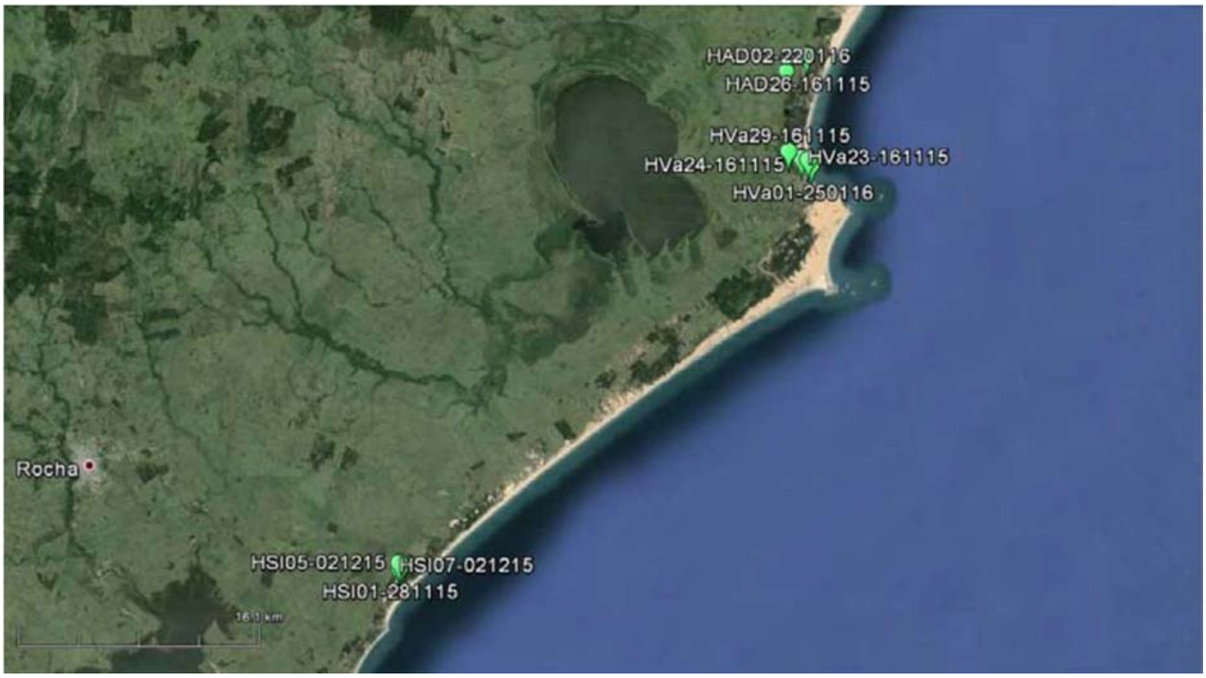

FIG. 1. Location of the sampling points

3. RESULTS AND DISCUSSION

The Atlantic coastal resorts of Rocha were chosen on the basis of three parameters: population, use of the water, and location relative to the black sand deposit. Samples were collected where the local population was greater than 100 but in summer grows to three or four thousand people. The black sand deposit is located between Barra de Valizas and Aguas Dulces, but mainly in the Barra de Valizas area. Santa Isabel de la Pedrera was also sampled because, although not immediately above the deposit, it is in its influence area. Importantly, groundwater is the main source of drinking water in these three resorts.

The measured gross alpha and gross beta activity concentrations are shown in Table 2. As expected, significant gross beta activities were found in Barra de Valizas, a resort which is located mostly above the black sand deposit. Significant gross alpha activities (the only ones above the minimum detectable activity) were found in Santa Isabel de la Pedrera, but there were also some low levels of activity. All the groundwater activity concentrations were in compliance with the Uruguayan standard for drinking water (combined gross alpha and gross beta activity <1 Bq/L) [10]. The World Health Organization guidelines for drinking-water [8] specify criteria of 0.5 Bq/L or less for gross alpha activity and 1 Bq/L or less for gross beta activity. These guidelines are consistent with a maximum effective ingestion dose of 0.1 mSv/a. All the groundwater samples were found to be within these guideline criteria and therefore radiologically suitable for drinking.

TABLE 2. RADIOACTIVITY IN GROUNDWATER

Sample code	Gross alpha activity (Bq/L)	Gross beta activity (Bq/L)
HAD02-220116	–0.00055 ± 0.00196	0.03842 ± 0.01386
HAD26-161115	0.00055 ± 0.00256	0.02903 ± 0.01291
HSI01-281115	0.01398 ± 0.00783	0.09006 ± 0.02059
HSI05-021215	0.00053 ± 0.00218	0.04588 ± 0.01475
HSI07-021215	0.01274 ± 0.00700	0.08005 ± 0.01929
HVa01-250116	–0.00078 ± 0.00179	0.09144 ± 0.01916
HVa23-161115	0.00109 ± 0.00224	0.19868 ± 0.02750
HVa24-161115	0.00109 ± 0.00216	0.18634 ± 0.02671
HVa29-161115	–0.00100 ± 0.00163	0.14817 ± 0.02394

Note: Concentrations exceeding the average of all the measured samples are underlined.

4. CONCLUSIONS

Samples taken from Barra de Valizas exhibited elevated values of gross beta activity concentration, while samples taken from Santa Isabel de la Pedrera exhibited elevated values of gross alpha activity. However, all the activity concentrations were within the applicable Uruguayan standards and World Health Organization guideline criteria. Further work is needed to determine gamma dose rates, to identify individual radionuclides in the water and to determine transfer factors between black sand and water as well as between water and living beings.

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Poster Presentation

HIGH PURITY GERMANIUM GAMMA-RAY DETECTOR FOR GROSS ALPHA AND GROSS BETA MEASUREMENTS IN WATER SAMPLES

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Abstract

The drinking water guidelines proposed by the World Health Organization and adopted by many countries entail the evaluation of gross alpha and gross beta activities in water. For this purpose, a method was developed in the laboratory, comprising a combined protocol involving alpha counting through a surface barrier detector and gamma ray spectrometry with an NaI(Tl) scintillation detector. Despite the reliability and usefulness of the method, it has some disadvantages, in that it involves the adoption of non-simultaneous readings and requires the evaporation of the water and counting of the alpha particles in the residue deposited on a planchet or disc, a procedure that may be affected by the sample salinity. A novel approach is described in this paper employing an EG&G ORTEC high purity germanium gamma ray detector, which was first calibrated for energy using the artificial radionuclides ^{137}Cs, ^{60}Co and ^{133}B as radioactive sources. Then, it was calibrated for concentration and activity concentration using various radioactive sources, with the efficiency of the gamma spectrometer being evaluated through these standards. The protocol adopted allowed the determination of gross alpha and gross beta activities in groundwater samples without the constraints of the previous technique.

1. INTRODUCTION

The current population growth has created an intense increase in the exploration of groundwater and surface water for human, agricultural and industrial purposes, where the need for quality control of drinking water is extremely important to assure the safety of the population. Radionuclides of natural origin affect the radiological quality of groundwater as a consequence of the geological–geochemical context of the aquifer systems. The World Health Organization has recommended screening tests for evaluating gross alpha and gross beta activities in water, suggesting that only when the screening criteria (gross alpha = 0.5 Bq/L, gross beta = 1 Bq/L) are exceeded, is it necessary to determine the radionuclide content using more sophisticated and time consuming procedures [1]. Many methodologies and detectors have been used to measure the gross alpha and beta activities in water over the last decades, among them the technique described in Ref. [2] that combines alpha spectrometry with Si(Au) surface barrier detectors and gamma ray spectrometry with an NaI(Tl) scintillation detector. However, the procedure has some disadvantages such as the non-simultaneous readings of each sample and alpha particle absorption on the dry residue generated after evaporation of the water sample. Semiconductor materials developed during the 1960s have now been applied to the detection of gamma rays associated with alpha and beta disintegration. In this regard, the high purity germanium (HPGe) detector now provides superior spectral resolution when compared with other detectors [3]. This paper describes the calibration steps of an HPGe gamma-ray detector as a simultaneous analyser of gross alpha and gross beta activities in water and its use for evaluating more than 100 water samples gathered from various aquifer systems in the Brazilian states of São Paulo, Minas Gerais and Mato Grosso do Sul.

2. MATERIALS AND METHODS

Three types of calibration were carried out:

(i) The spectrometric system was first calibrated for energy using four different gamma-ray photopeaks from radioactive sources containing the artificial radionuclides ^{137}Cs, ^{60}Co and ^{133}Ba. A plot of energy (in keV) versus channel number was obtained over a wide spectrum of gamma energies.

(ii) Certified standards provided by the New Brunswick Laboratory of the US Department of Energy, Argonne, IL, consisting of pitchblende and monazite sand with different uranium and thorium concentrations, were utilized for the calibration for uranium and thorium concentration. Pure KCl and different mixtures of this matrix with SiO_2 were also used to calibrate for potassium concentration. Eight gamma ray photopeaks were chosen from the ^{238}U and ^{232}Th decay series [4]:
   - ^{238}U decay series: ^{214}Bi (1120.19 and 1764.49 keV, beta decay) and ^{226}Ra (186.1 keV, alpha decay);
   - ^{232}Th decay series: ^{208}Tl (583.19 and 2614.53 keV, beta decay), ^{228}Ac (911.21 and 968.97 keV, beta decay) and ^{224}Ra (240.9 keV alpha decay).

   The two alpha emitting radionuclides (^{224}Ra and ^{226}Ra) were chosen despite overlapping with or being very close to other gamma ray photopeaks, in the absence of more isolated viable peaks. For potassium, the 1461 keV electron capture photopeak for ^{40}K was used. Several plots were generated of concentration (in parts per million) versus effective intensity (the counting rate in counts per second, per unit mass in grams).

(iii) The last calibration was done for activity concentration (in Bq/g), correlating the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K with the effective intensity, using the same photopeaks as in (ii) above.

In all readings, the background activity was measured and subtracted from the gamma spectrum of each standard. Subsequently, a percentage efficiency detection curve was generated as a function of the gamma ray energy according to Eq. (1).

$$\varepsilon f = \frac{cr - BG}{f \times A} \tag{1}$$

where εf is the detection efficiency (%) for the gamma ray energy of a specific radionuclide, cr represents the measured counting rate (counts per second), BG is the background counting rate (counts per second), f is the peak intensity (%) and A is the known standard activity. In this case, additional gamma ray photopeaks were adopted, i.e. ^{210}Pb (46.54 keV), ^{214}Pb (242, 352 and 295.2 keV) and ^{214}Bi (609 and 2204.21 keV). The efficiency is a factor directly dependent on the geometry of the detector–sample system [5].

The groundwater samples to be analysed were collected from water wells and pipes located in various municipalities in the Brazilian states of São Paulo, Minas Gerais and Mato Grosso do Sul and stored in sealed polyethylene containers. The analyses were done using an EG&G ORTEC gamma spectrometric system with a coaxial HPGe detector and Gamma Vision Software installed at LARINIonizing Radiation Laboratory at Instituto de Geociências e Ciências Exatas,Universidade Estadual Paulista em Rio Clara (IGCE-UNESP-Rio Claro). The reading for each sample lasted at least 30 000 s (~8 h) using a 1 L capacity Marinelli container. The calibration curves were used to calculate the alpha and beta activities of the samples, taking into account the separation of the superimposed alpha related gamma-ray peaks, according to Eq. (2).

$$A = \frac{cr - BG}{f \times \varepsilon f} \quad (2)$$

The activity value obtained was divided by the sample volume V to yield the activity concentration As of the water sample.

$$As = \frac{A}{V} \quad (3)$$

The final gross beta activity was calculated by the sum of the averages of partial specific activities of ^{40}K and each representative radionuclide from the uranium and thorium decay series, whereas the gross alpha activity was obtained by the ^{226}Ra and ^{224}Ra decay peaks belonging to the ^{238}U and ^{232}Th decay series, respectively.

3. RESULTS AND DISCUSSION

The result of the energy calibration is shown in Fig. 1. Examples of the results of the calibrations for concentration (in parts per million) and activity concentration (in becquerels per gram) are shown in Figs 2–4 and 5–7, respectively.

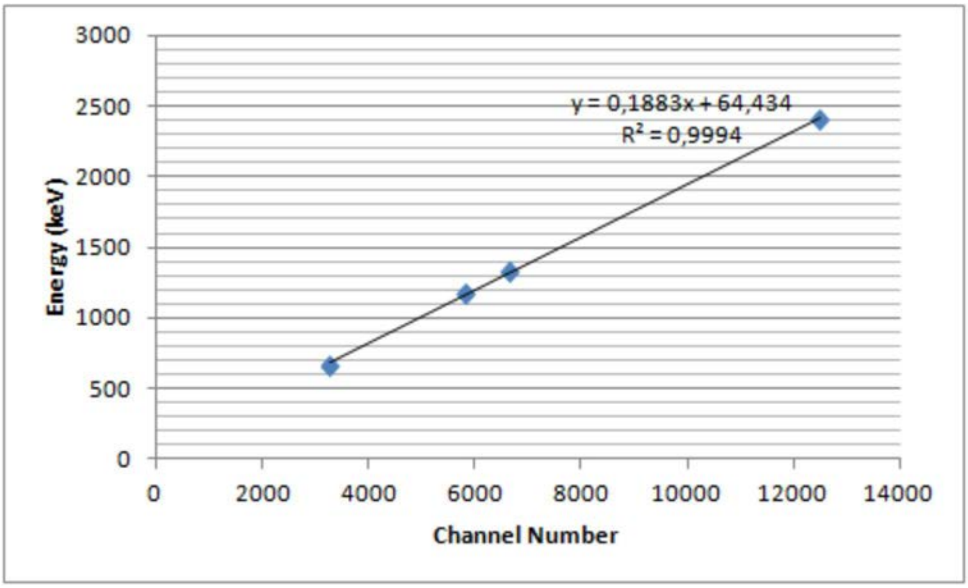

FIG. 1. Calibration for energy

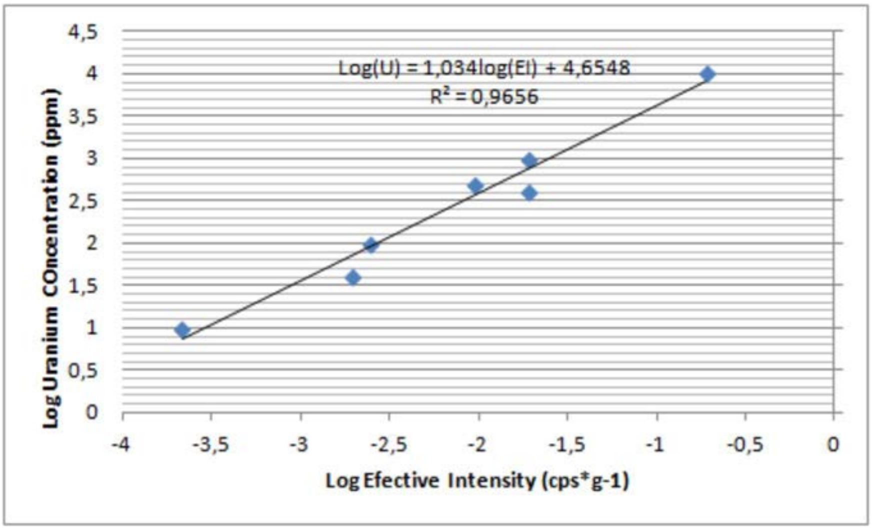

FIG. 2. Calibration for uranium concentration.

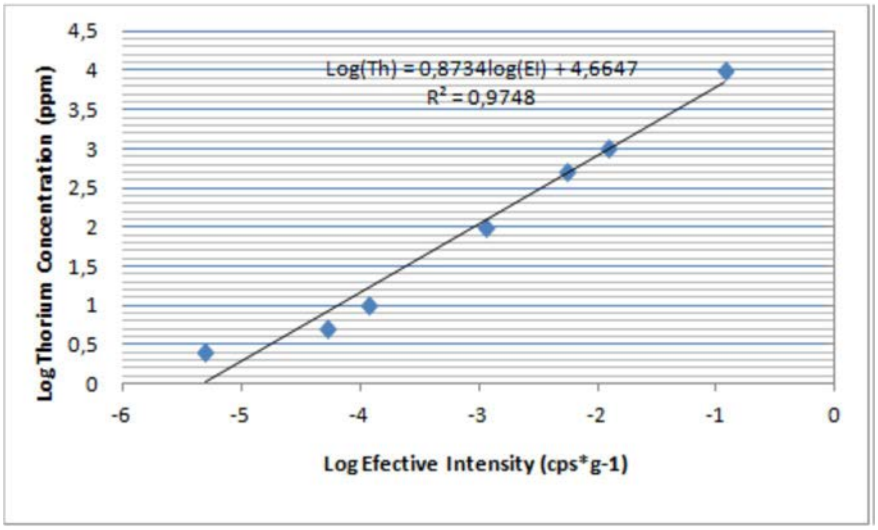

FIG. 3. Calibration for thorium concentration.

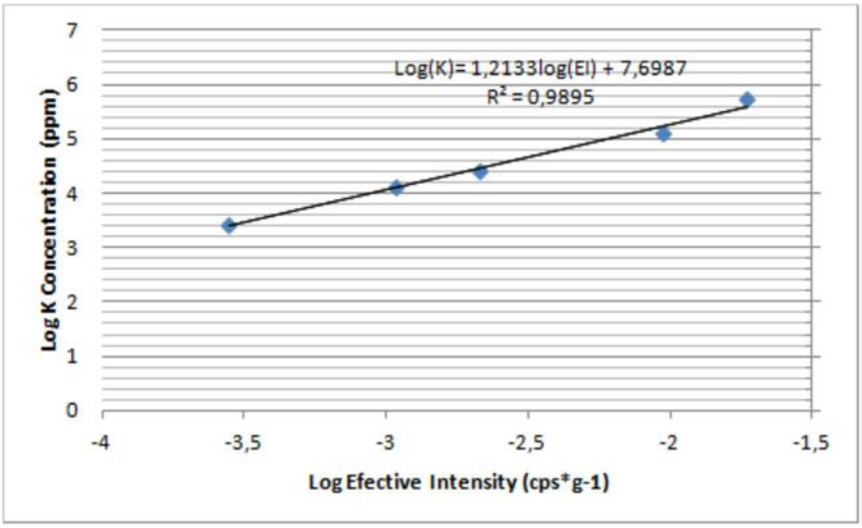

FIG. 4. Calibration for potassium concentration.

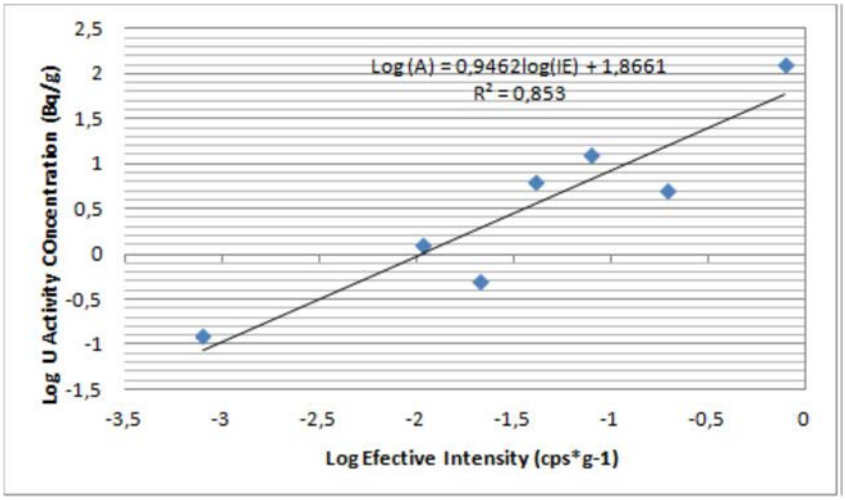

FIG. 5. Calibration for uranium activity concentration.

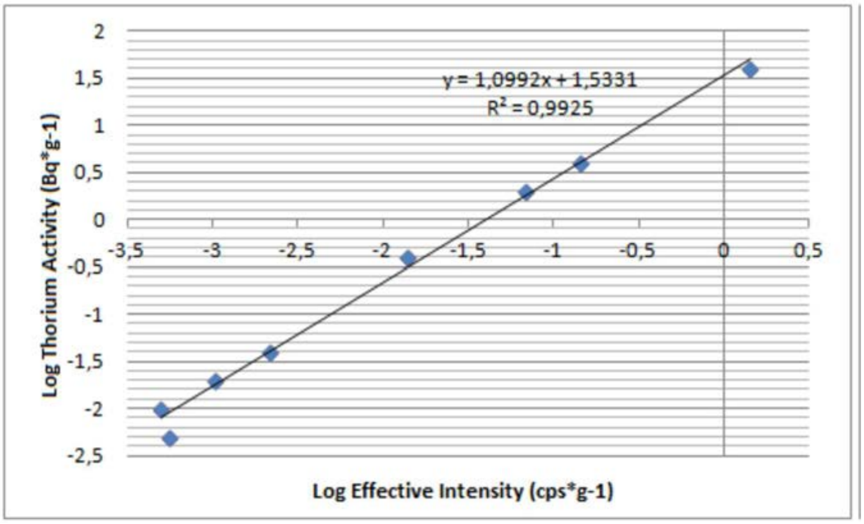

FIG. 6. Calibration for thorium activity concentration.

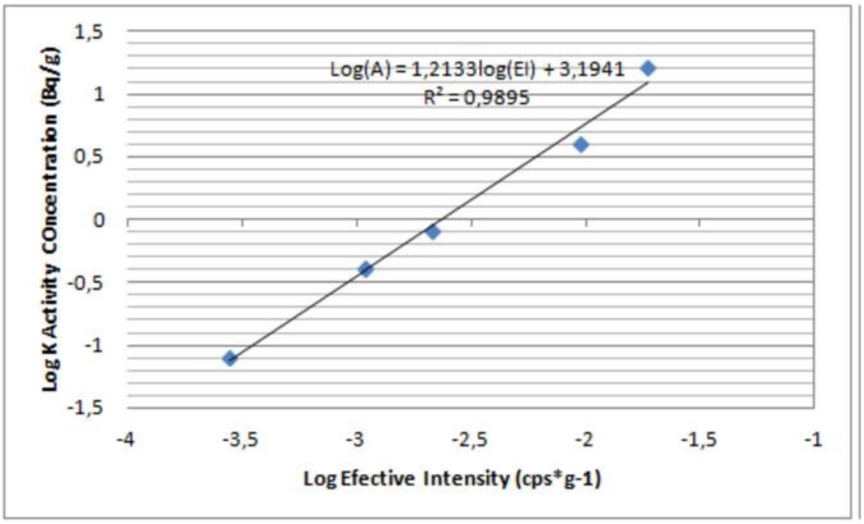

FIG. 7. Calibration for potassium activity concentration

The results for the detection efficiency were represented by two separate polynomial functions, the first covering lower gamma ray energy levels (from 46 to about 1000 keV) and the second covering energies from 1000 to 2600 keV. The general trend, shown in Fig. 8, indicates that higher gamma ray energy values corresponded to lower detection efficiency levels, as expected.

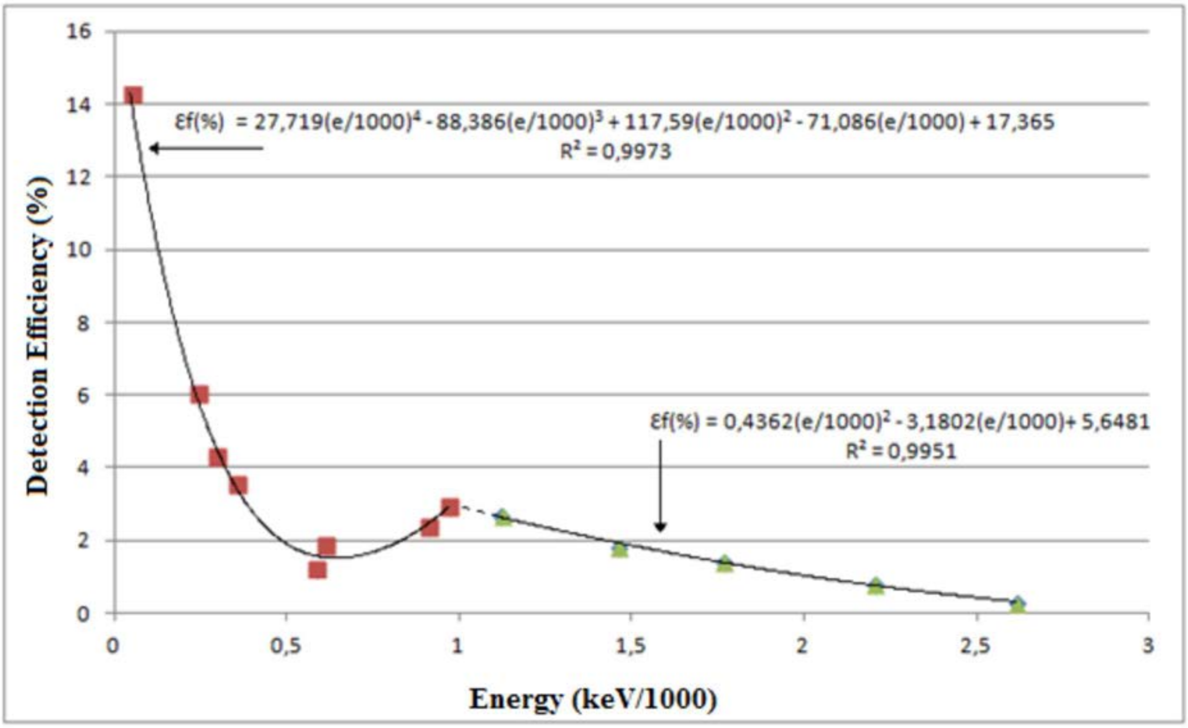

FIG. 8. Detection efficiency represented by two polynomial functions.

The final gross beta activity results obtained through ^{208}Tl were significantly higher than those obtained through ^{228}Ac, contrary to expectations. The World Health Organization guideline criteria [1] were exceeded by a significant number of groundwater samples, but especially so when using the ^{208}Tl calibration procedure — about 77% of the samples showed activities above 1 Bq/L whereas, using the ^{208}Ac calibration procedure, 80% of them were below the criteria. In general, water from fractured aquifers from Caxambu, Cambuquira, São Lourenço, Lambari, Poços de Caldas and Serra Negra municipalities exhibited higher gross beta activity values. Practically all the samples resulted in gross alpha activities above 0.5 Bq/L, with large contributions from ^{224}Ra and, again, the most significant values were associated with those same municipalities and fractured aquifers. In some samples, the activity of ^{226}Ra was so low that the ^{226}Ra–^{235}U photopeak was barely identifiable in the gamma spectrum, especially for the samples collected in the Paraná Basin aquifer systems, where comparatively low radioactivity levels could be expected.

The difference between the results calculated through ^{208}Tl and those calculated through ^{228}Ac could be explained by the type of efficiency curve used and/or by a deviation from equilibrium conditions as a result of the escape of radon gas from the radioactive sources and samples. Results calculated through ^{208}Tl would be affected by the escape of radon, whereas results based on the use of a 'pre-radon' radionuclide belonging to the Th decay series (^{228}Ac) would not be affected. The geological and geochemical characteristics of the aquifers are certainly the main factors affecting the radioactivity levels in the groundwater. The cities with relatively high gross alpha and beta activities have their groundwater associated with orthogneiss, migmatites, garnet schists, pegmatites, and intrusive alkaline rocks [6]. In contrast, water samples from the Paraná Basin showed lower gross alpha and beta activity results, as would be expected from the geology-related factors. The behaviour of radionuclides of natural origin under different geological and geochemical conditions may explain the gross alpha and beta values. However, they should be better investigated considering the mobility and/or solubility of uranium, thorium and their progeny and taking into account anthropogenic inputs such as agrochemical products and/or industrial contamination.

4. CONCLUSIONS

The results verified the viability of the proposed technique as an alternative to the previous method developed for gross alpha and gross beta radioactivity determination. It allows water samples to be analysed without the use of chemicals, sample destruction, problems caused by salinity or even the adoption of more than one spectrometric system for the data acquisition. Shortcomings of the technique can be attributed to a lack of isolated and intense photopeaks for alpha calibration and also to potential problems caused by non-equilibrium conditions resulting from to the escape of radon gas. In this case, the use of a ‘pre-radon’ radionuclide for calibration, such as ^{228}Ac in the Th series, is highly recommended. Some of the groundwater samples indicating high gross alpha and beta activity values could be re-evaluated considering seasonal changes or even possible anthropogenic inputs perhaps associated with agriculture.

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Poster Presentation

NaI(Tl) DETECTORS FOR ENVIRONMENTAL GAMMA SURVEY AND BACKGROUND RADIATION MAPPING OF VENUES AND OTHER STRATEGIC LOCATIONS FOR MAJOR PUBLIC EVENTS

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Abstract

The original radiometric pattern due the presence of radionuclides of natural origin is highly altered by the use of decorative stones and building material and also by new concrete structures and highways in urban areas. One of the most versatile classes of mobile radiation detection instruments for the identification of radiation sources is the gamma spectrometer. Some units also have identification software, GPS capability and even Wifi or GSM functionalities. NaI(Tl) detectors with these features are very useful for environmental surveys and for nuclear security systems and measures for major public events. The former involves the gathering of information about the normal background, concentrations of radionuclides of natural origin, and radiometric mapping. The latter, as suggested in IAEA Nuclear Security Series No 18, can be performed before a major public event for the detection of unauthorized nuclear and other radioactive material and for determining a baseline radiation level should a nuclear security incident occur.

1. INTRODUCTION

In the last decade, several major public events have taken place in Rio de Janeiro: the FIFA Confederations Cup in 2013, the FIFA World Cup in 2014 and the Rio Olympics in 2016. The threat of nuclear and radiological terrorism should not be ignored at such events. The greater the damage, the lower should be the possibility of occurrence. To reduce this risk, the National Commission on Nuclear Energy (CNEN) and its Radiation Protection and Dosimetry Institute (IRD) adopt strong nuclear security procedures [1, 2], systems and measures including technical cooperation, training courses, donations and equipment loan, and the presence of foreign experts in the pre-event stages and even during events. Seminars and conferences were held in the past ten years since the Pan American Games in 2007. The IAEA and the US Department of Energy worked in deep collaboration with IRD and CNEN to meet the relevant nuclear security responsibilities [2]. Different models of detectors can be used for detection of radioactive material in the form of natural, medical or industrial sources, but for criminal or unauthorized acts involving nuclear or other radioactive material different approaches should be used [1]. One of the most important tasks to be performed before the event is the background radiation mapping of venues, serving as a baseline for the natural background level [2]. For this task, the best option is the use of gamma spectrometers with georeferencing capabilities [3–6].

2. METHODS

The environmental radiation pattern is too complex to be recreated in the laboratory [7]. Generally detectors used for dose rate measurements are calibrated using Monte Carlo techniques and sources such as ^{137}Cs (667 keV) or ^{60}Co (1.17 MeV and 1.33 MeV), but the energies associated with radionuclides in the ^{238}U and ^{232}Th series reach values of up to

2615 keV (^{208}Tl). The best way to calibrate these detectors for measuring H*(10) is using the own environmental field radiation. At IRD there are large planar sources (known as PADs) with specific concentrations of U_{nat}, Th_{nat}, ^{40}K, mixtures of uranium–thorium and background reference material [6, 8]. The detectors used in this work, Atomtex AT6101c[1], were stabilized and checked against these planar sources (see Figs 1 and 2). Also the detectors were checked against ^{226}Ra, ^{137}Cs and ^{60}Co test sources and the results were compared with those from other detectors and from another unit of the same type of detector. After the stabilization, this detector was used to measure H*(10). The results can be displayed on any computer that runs the software Google Maps®. The pathway, data, coordinates, time stamp, radionuclides identified and dose rate (H*(10)) are shown on the computer display.

FIG. 1. Detector AT6101c in one of the planar sources of IRD.

FIG. 2. E 01: AT6101c IdentiFINDER® 2; E 02: Exploranium GR130; D 01: comparison between two AT6101c units; D 02: agreement between the GR130 and AT6101c at low background measurements. All the units are miocrosieverts per hour except the GR130 that displays in units of nanosieverts pere hour.

3. RESULTS

Values of H*(10) for the Olympic Stadium Nilton Santos at Rio de Janeiro are shown in Fig. 3. The statistical analysis of this set of data shows 606 spectra collected (10 s each), a mean H*(10) of 190 nSv/h and a standard deviation of 38 nSv/h. The minimum and maximum values

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[1] http://www.atomtex.com/sites/default/files/at6101c_0.pdf.

are 103 and 351 nSv/h, respectively. The values extend beyond normal background values, but it should be noted that the stadium is not a natural environment. Also the values are in accordance with previous work [4–6].

FIG. 3. Results of a survey at Olympic Stadium Nilton Santos (Engenhão). All the results are in nanopsieverts per hour. The white dots are the lower values and red dots are the higher values in this area.

4. DISCUSSION

During the preparatory period for the 2016 Rio Olympics, a massive change occurred in the city. Viaducts, tunnels, bridges and large condominiums were built to improve urban mobility and host the ‘Olympic family’ (see Fig. 4).

In urban areas the dose rate depends on the type of material (e.g. concrete, asphalt, bare earth) and vegetation and the presence or absence of artificial radioisotopes, whether by accident, use of radiopharmaceuticals, discharge, fall-out or release into the environment. The results obtained in an urban area should be analysed in conjunction with several other factors, because the presence of different building materials may change the dose rate of a particular place, either by increasing the value after the addition of material with higher content of radioactive material or decreasing as a result of the presence of shielding material. A large quantity of concrete was used over areas with a low natural radioactive background. At some locations, the new background of the venues was high enough to trigger the alarm of detectors used for access control. This increase in the background was detected by the AT6101c detector, or IdentiFINDER, and Exploranium GR130. The survey areas submitted to radiometric scanning must be well characterized because even in visually identical compartments such as sand beaches, dose rates may range from 30–35 nSv to 300 nSv/h (in the presence of monazite sands) owing to the chemical composition of the sand. The values for water bodies vary from 10 to 20 nSv/h and for land measurements we can find values ranging from as low as 30–40 nSv/h in lateritic soils from the Brazilian Midwest up to 200 nSv/h in granitic pavements or mountains. [4–6].

FIG. 4. Environmental alterations near the Olympic Village (indicated by the red arrow), showing the infrastructural alterations for transport (Transolímpica Highway) and the buildings of the Olympic Village.

5. CONCLUSIONS

Gamma spectrometers are very suitable for use in environmental surveys. They are capable of detecting and identifying radioisotopes and also register the dose rate H*(10). The capability of producing data with geographical coordinates, time stamp and radioisotopes identified allows the operator to produce maps and display the results in a few minutes as the software generates, without further statistical analysis, the data being displayed using the free software Google Earth. The size of the files, with the extension ".kml", is very small, allowing them to be transmitted even via weak internet connections, generating a rapid response for the security systems. The stabilization and checking of this detector should be carried out carefully because, even when the dose rate is shown, the identification process will be compromised if the stabilization fails. Major public events provide a very good opportunity for training and verification, in real conditions, the response of instruments and persons. Most of the gamma radiation alarms at the venues were from radionuclides of natural origin (isotopes of potassium, uranium and thorium) and medical isotopes (^{99}Tc, ^{131}I being the most common). In portals with X ray monitors or near mobile X ray machine detectors, the AT6101c is also capable of detecting X ray radiation through the indication of bremsstrahlung.

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Poster Presentation

RADON ACTIVITY CONCENTRATIONS IN AN EXPERIMENTAL LIVING ENVIRONMENT IN CURITIBA, BRAZIL

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Abstract

Radon gas and its short half-life progeny are responsible for about half of the radiation dose received by individuals. The inhalation of radon is considered to be the second leading cause of lung cancer. In Brazil, the regulatory body has established an annual dose limit of 10 mSv, consistent with an indoor radon concentration of about 300 Bq/m^3. Radon concentrations were measured indoors (in two prefabricated houses) and outdoors, using 40 CR-39 detectors installed in diffusion chambers. The houses were built in an experimental environment, on the premises of the Applied Nuclear Physics Laboratory (UTFPR)/Ecoville, located in the city of Curitiba, Paraná. House 1 was built using construction material from the Czech Republic, while House 2 was built using material from Brazil. Following a three month exposure period, the detectors were etched and the radon concentrations obtained from the calibration methodology developed by UTFPR and the Nuclear Technology Development Centre (CDTN). All the activity concentrations were found to be below 300 Bq/m^3. The average indoor concentrations were 10.1 ± 1.3 and 7.8 ± 1.1 Bq/m^3 for House 1 and House 2, respectively, while the outdoor radon concentration was found to be 11.5 ± 2.9 Bq/m^3. On the basis of these results, no remedial action would have been necessary.

1. INTRODUCTION

Radon is a gas that occurs naturally in the form of the two main radioisotopes ^{222}Rn (half-life 3.8 d) and ^{220}Rn (half-life 55 s) [1]. It emanates from the ground in all directions. Concentrations of radon are low in the open air because of continuous dispersion, but can be 2–20 times higher in indoor environments. In Brazil, the regulatory body has established an annual dose limit of 10 mSv, consistent with an indoor radon concentration of about 300 Bq/m^3 [2, 3]. When inhaled, radon decays within the lung, and the subsequent decay of its short-lived progeny leads to an increased risk of lung cancer. The associated probability depends on the exposure time and the radon concentration. For high concentrations of radon, remedial measures are justified and recommended [1]. In this study, radon activity concentrations were measured indoors (in two prefabricated houses) and outdoors.

2. METHODS

The solid state detector used in this study was made from a polymer called Columbia Resin 39 (CR-39). It is sensitive to alpha radiation, is readily obtainable and easy to install, and exhibits optimum contrast for the visualization of nuclear tracks [4]. The detector chamber is designed to promote greater homogeneity in the process of detection and to reduce the deposition of ^{222}Rn decay products. The lid of the chamber allows the passage of air to its interior. A borosilicate filter is located between the cover and the detector. The preparation of detectors began with the cleaning of the diffusion chambers followed by assembly as shown in Fig. 1.

FIG. 1. Detector assembly sequence. A: the filter is placed in the chamber; B: the detector is added; C: the assembly is mounted and the chamber numbered.

Forty detectors were exposed inside and outside two prefabricated houses experimentally constructed at the Applied Nuclear Physics Laboratory (UTFPR) at the Ecoville headquarters, located in Curitiba, Paraná. One of these houses was built with construction material from the Czech Republic and the other with material from Brazil. The exposure time was approximately three months, sufficient to provide the necessary data while preventing overlapping traces. After the exposure period, the detectors were chemically etched with 6.5M NaOH solution for 14 h. Counting of the nuclear tracks was carried out manually using an optical microscope. The radon concentrations were calculated in accordance with the calibration methodology developed by the Nuclear Technology Development Center (CDTN) in cooperation with UTFPR [5]. The calibration factor relates the track density and exposure time to the ^{222}Rn concentration. The results were evaluated against the recommendations of the International Commission on Radiological Protection [1].

3. RESULTS

The results are shown in Tables 1–3. The radon concentrations were all below the 300 Bq/m^3 reference level.

TABLE 1. RADON CONCENTRATIONS IN HOUSE 1

Location	Concentration (Bq/m^3)
Kitchen drawer	12.5 ± 0.9
Bathroom shower	7.7 ± 0.6
Air extractor	5.2 ± 0.4
Bathroom towel rack	15.0 ± 1.1
Small table in room	13.6 ± 1.0
Furniture above refrigerator	8.0 ± 0.6
Floor	3.0 ± 0.2
Inside flowerpot	22.7 ± 1.7
Kitchen accessory	8.0 ± 0.6
Shelf above refrigerator	7.5 ± 0.6
Stool close to wall	7.5 ± 0.6
Above cupboard cups	8.6 ± 0.6
Sofa foot	8.2 ± 0.6
Access hole under kitchen oven	16.1 ± 1.2
Wardrobe	3.9 ± 0.3
Near house entrance	14.5 ± 1.1
Average	10.1 ± 1.5

TABLE 2. RADON CONCENTRATIONS IN HOUSE 2

Location	Concentration (Bq/m^3)
Drawer from double bedroom	6.6 ± 0.5
Base of kitchen cabinet	14.3 ± 1.1
Pillow in single bedroom	2.0 ± 0.2
Under kitchen table	0.0 ± 0.0
Double bedroom ceiling	13.6 ± 1.0
Top shelf of kitchen cupboard	15.5 ± 1.1
Bottom shelf of kitchen cupboard	11.4 ± 0.8
On kitchen shelf	5.2 ± 0.4
Behind stove	6.1 ± 0.5
Behind bathroom wash basin	5.5 ± 0.4
Inside chest	0.9 ± 0.1
Toilet paper cover	0.7 ± 0.1
Above wardrobe in single room	8.9 ±0.7
Amplifier under television	7.3 ± 0.5
Desk	6.8 ± 0.5
Outside washing tank	12.7 ± 0.9
Laundry tank	4.5 ± 0.3
Near entrance to house	12.7 ± 0.9
Over lamp in laundry	12.5 ± 0.9
Average	7.8 ± 1.2

TABLE 3. OUTDOOR RADON CONCENTRATIONS

Location	Concentration (Bq/m^3)
Water register at external door	15.9 ± 13.6
Rainwater pipe collector	13.6 ± 1.0
Plant next to entrance	18.2 1.3
Garden next to bench	2.5 ± 0.2
Metal pillars in garage	7.3 ± 0.5
Average	11.5 ± 3.0

4. CONCLUSION

Since the radon concentrations were far below the reference level of 300 Bq/m^3 (and, indeed, less than the average concentration found in homes in the city of Curitiba [5, 6]), no remedial action would have needed to be considered. Nevertheless, despite the growing interest in the subject shown by several studies that have emerged in Brazil, there should be greater incentives given by the government and also a greater awareness of the public about the risk associated with prolonged exposure to radon.

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Poster Presentation

VARIABILITY AND SPATIAL DISTRIBUTION OF RADIONUCLIDES OF NATURAL ORIGIN IN THE STATE OF MINAS GERAIS

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Abstract

The study area comprised the whole territory of the state of Minas Gerais, which has unique characteristics in terms of lithology, genesis and morphology of soils, having a major influence on the pedogeochemistry of trace elements, metals and radionuclides. From the results of activity concentration measurements of ^{238}U ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra in 153 samples randomly collected throughout the state, statistical studies were conducted to assess their variability and spatial distribution. For this purpose, appropriate statistical tools such as control charts and GIS were used. The application of control charts allowed the results to be arranged in such a way as to verify the presence of trends and outliers. The activity concentrations of radionuclides in the ^{238}U and ^{232}Th series varied significantly, confirming the great diversity and heterogeneity of natural radioactivity present in the soil. It was also observed that most results were within the expected statistical limits. The activity concentrations of the radionuclides considered as outliers were observed in regions of Minas Gerais where there are radioactive anomalies.

1. INTRODUCTION

The existence of a contaminated area can lead to problems such as health risks, impaired quality of water resources, restrictions on land use and environmental damage. With the aim of streamlining the decision making process in the management of contaminated areas, setting priorities and reducing costs, environmental agencies of various countries, including Brazil, have chosen to establish guidance values of soil quality, to be used at the initial stage of the decision process. A quality reference value (QRV) is defined as the concentration of a substance in soil or groundwater that can be used for soil to be deemed as being clean or for groundwater to be deemed as being of natural quality. However, it should be noted that this practice can lead to inappropriate ratings, since there are striking differences in the physical characteristics of each region, in particular the geological conditions, the water and the soil. These differences justify the development of a proper table of QRVs for natural radionuclides in specific soils for each region. The area covered by this study is the state of Minas Gerais. Located in south-eastern Brazil, Minas Gerais is the fourth largest state in surface area, occupying an area of 588 384 km^2 (7% of Brazilian territory) and representing the third largest economy of the country. Minas Gerais extracts more than 160 million t/a of iron ore, and accounts for 29% of national mineral production, 53% of metallic minerals production and about 50% of gold production[1]. The mining and processing of ores containing, among others, tin, uranium, phosphate, niobium and rare earths contribute to the generation of large amounts of NORM residues. The improper disposal of these residues can cause situations that result in soil and groundwater contamination and undue exposure of members of the public. Thus, studies that contribute to the establishment of criteria and guidance values for the prevention and control of potentially toxic chemicals in soil and groundwater are of fundamental importance for the management of contaminated areas in the State of Minas Gerais.

2. METHODS

The study area covers the entire state area, which has unique characteristics in terms of lithology, genesis and morphology of soils, having a major influence on the pedogeochemistry of trace elements, metals and radionuclides. One hundred and fifty three soil samples were randomly collected at a depth of 0–20 cm within the mapping unit, observing the predominance of vegetation preserved or minimally impacted. The uranium and thorium concentrations present in the soil samples were determined by mass spectrometry by inductively coupled plasma (ICP-MS). All analyses were performed in the laboratory SGS GEOSOL, using as the separation method fusion with lithium metaborate [1]. This is one of the most widely used methods for the determination of trace elements in soil because of advantages such as multi-element capacity and low detection limits. For the analysis of ^{226}Ra, ^{210}Pb and ^{228}Ra, gamma spectrometry was used and in all samples semi-quantitative X ray determinations were performed for macroelements in order to take into account the dependency of self-absorption on the macroconstituents. The QRV of each substance was based on the 75th percentile of the sample population and was determined using relevant statistical analysis and in accordance with the design of the sampling plan and the sample set obtained [2]. The ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra concentrations were subjected to statistical analysis to assess their variability and spatial distribution. Therefore, we opted for the application of control charts which allow the results to be arranged in such a way as to verify the presence of trends and outliers. From the identification of sampling points that presented activity concentration values above the limits of the control charts (upper control limits), regional investigations to correlate them with some kind of anomaly or geological phenomenon were carried out. For this purpose, GIS tools were used.

3. RESULTS AND DISCUSSION

The control charts for each radionuclide are shown in Figs 1–5. In these charts, one can observe a very similar behaviour between the elements of the same number, i.e. the ^{238}U series (^{226}Ra, ^{210}Pb) and the ^{232}Th series (^{228}Ra). Figure 6 shows the locations of sampling sites exceeding the upper control limits for both series (^{238}U and ^{232}Th). The distribution parameters for the various radionuclides are presented in Table 1. The standard deviations were very high owing to the high variability of the results, leading to high values for the upper control limit. Tables 2 and 3 show the outliers found for each radionuclide of the ^{238}U and ^{232}Th series, respectively, its location and the main prospected minerals.

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[1] Statistics provided by Companhia de Desenvolvimento Econômico de Minas Gerais, 2012.

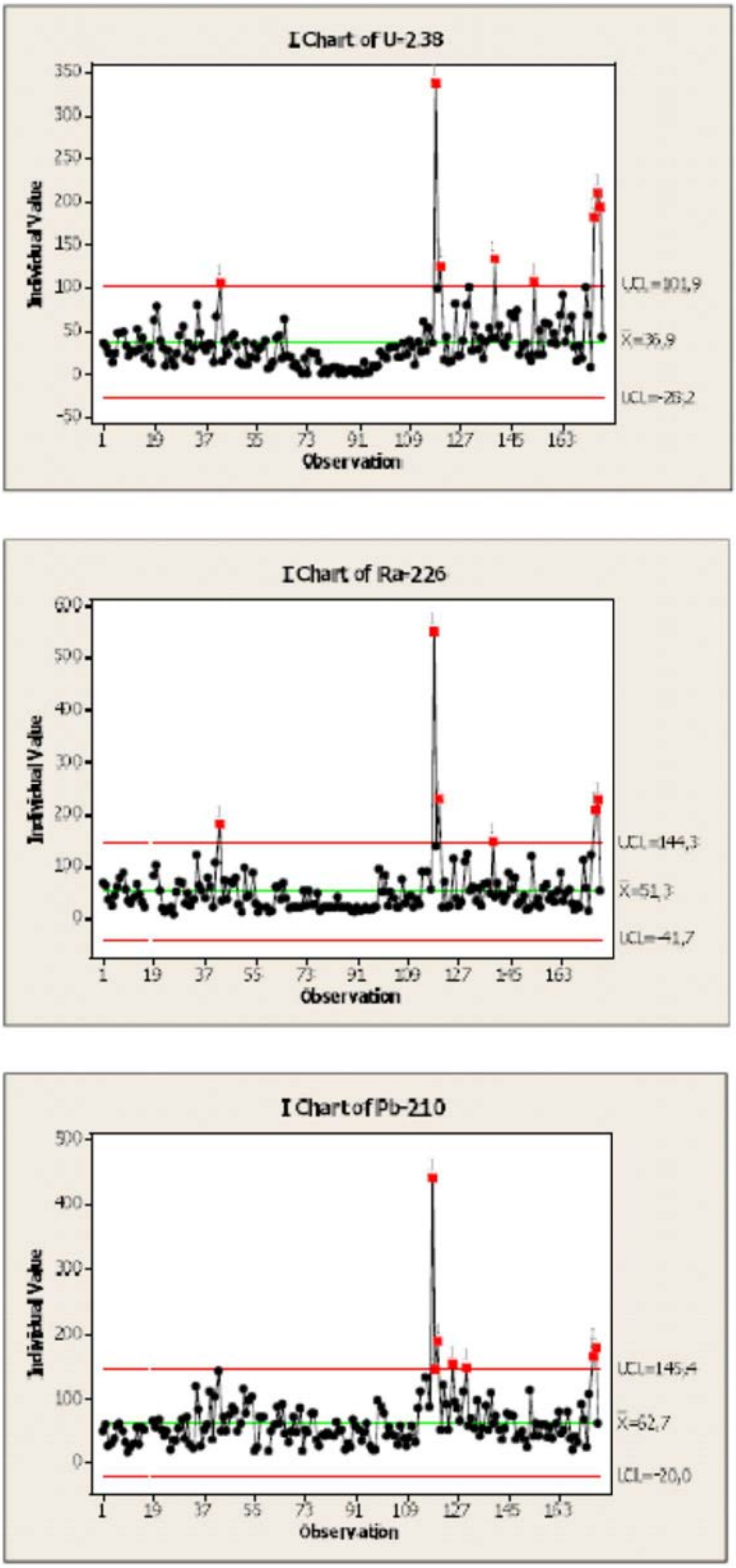

FIG. 1. Control chart for activity concentrations of ^{238}*U series radionuclides.*

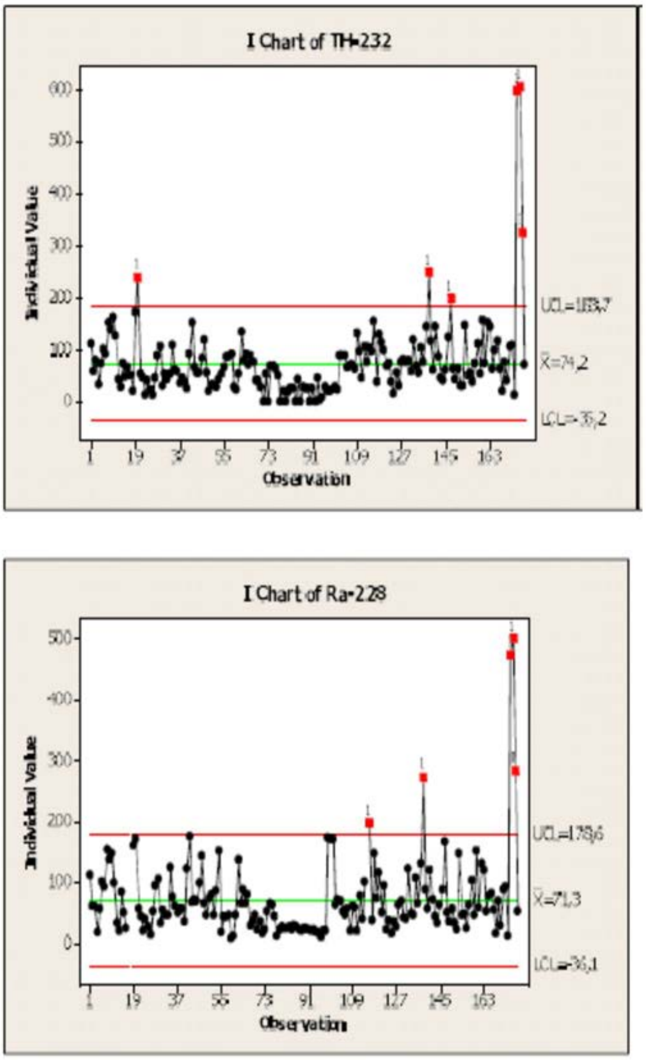

FIG. 2. Control chart for activity concentrations of ^{232}Th series radionuclides.

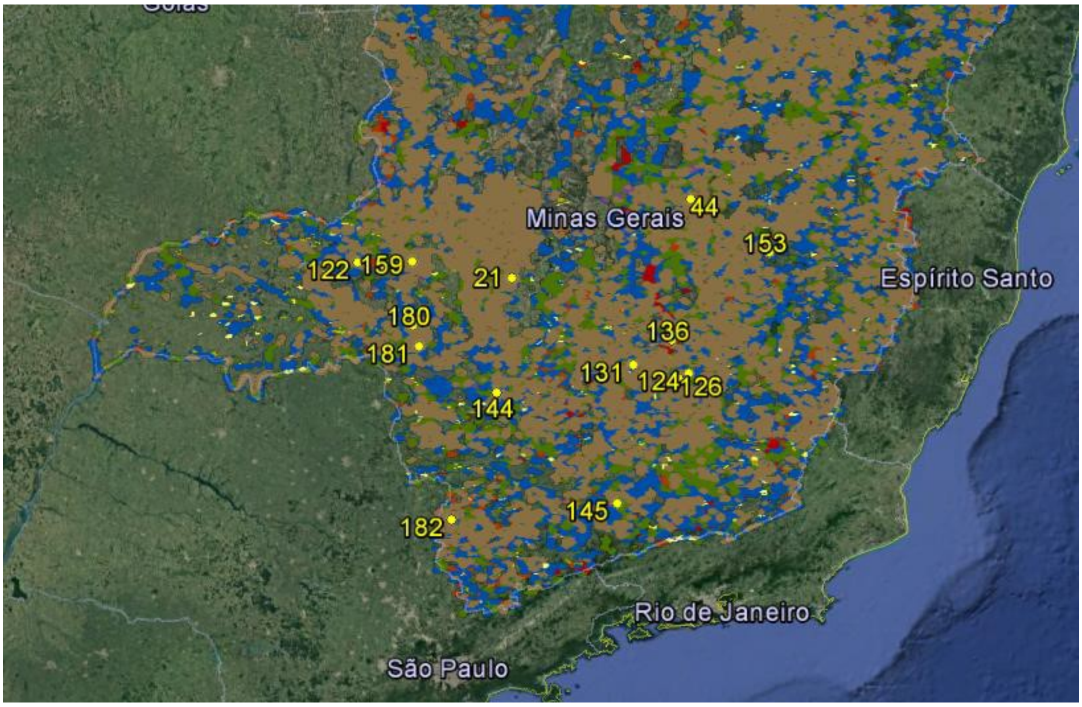

FIG. 3. Locations of outliers.

TABLE 1. PARAMETERS OF THE ACTIVITY CONCENTRATION DISTRIBUTIONS

	Activity concentration (Bq/kg)			
	QRV (75th percentile)	Mean	Standard deviation	Upper control limit
U-238	43.25	36.9	39.8	101.9
Ra-226	64.1	51.3	54.1	144.3
Pb-210	76.4	62.7	43.4	145.4
Th-232	90.8	74.3	74.6	183.7
Ra-224	88.1	71.3	65.9	178.6

TABLE 2. URANIUM-238 SERIES RADIONUCLIDES AT LOCATIONS CONSIDERED AS OUTLIERS

Sample no.	Activity concentration (Bq/kg)			Location	Major minerals
	U-238	Ra-226	Pb-210		
44	105	181	–	Diamentina	Gold, quartz
124	337.8	549.8	441.5	Ouro Preto	Iron, gold
126	125.3	229.6	188.9	Amarantina	Gold
131	–	–	153.5	Igarapé	Iron
136	–	–	147.5	Sabará	Iron, gold
145	134	–	–	São João del Rey	Iron
159	106.5	–	–	Patrocínio	Titanium, phosphate
180	181.9	–	–	Araxá	Niobium, phosphate
181	210.3	207	166	Araxá	Niobium, titanium, manganese
182	193.6	228	180	Poços de Caldas	Bauxite

TABLE 3. THORIUM-232 SERIES RADIONUCLIDES AT LOCATIONS CONSIDERED AS OUTLIERS

Sample no.	Activity concentration (Bq/kg)		Location	Major minerals
	Th-232	Ra-228		
21	239	–	Abaeté	Phosphate
122	–	198.3	Romaria	Gold, diamonds
144	249.7	272	Piumhi	Iron, copper, chromium, nickel, dolomite
153	199.7	–	Sapucaia de Guanhães	Gold, granite
180	597.9	474	Araxá	Niobium, phosphate
181	606	501	Araxá	Niobium, titanium, manganese
182	324.7	284	Poços de Caldas	Bauxite

4. CONCLUSIONS

The concentrations of ^{238}U and ^{232}Th series radionuclides varied significantly, confirming the great diversity and heterogeneity of Minas Gerais soils. However, most samples were within the expected statistical limits. It is noteworthy that the activity concentration values considered as outliers were observed in the regions of Minas Gerais where there are radioactive anomalies, such as the pegmatitic provinces in the north-east of the state, the alkaline magmatic provinces of Pocos de Caldas, the alkaline and basic intrusives in Tapira, Araxá and Salitre and the migmatite regions of São João del Rey and Itabirito, thus corroborating the results obtained in this study.

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Poster Presentation

RADIONUCLIDES OF NATURAL ORIGIN IN FOOD AND WATER FROM A HIGH BACKGROUND RADIATION AREA IN SOUTH-EASTERN BRAZIL

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Abstract

Food consumption is one of the main entry routes of radionuclides into the human body. Concentrations of radionuclides of natural origin vary according to several factors such as local geology, climate and agricultural practices. High background radiation areas have attracted much interest from a radiation protection point of view. The Poços de Caldas Plateau is located in south-eastern Brazil and is regarded as a high background radiation area. The situation of the resident population in the region of Poços de Caldas is of special interest for public health surveillance. The activity concentrations of radionuclides of natural origin in 19 groups of food and drinking water were determined. These radionuclides were quantified using gamma spectrometry, ultra low level alpha and beta total counting, and alpha spectrometry, after group preparation and/or radiochemical separations. The results of this study showed that the food groups analysed give rise to a low effective ingestion dose. Calculations were performed using the total radionuclide concentration in each food group, the mass consumed per day by each population group, the total time considered, and the dose intake of each radionuclide. The average annual effective dose received by members of the urban population on the Poços de Caldas plateau was 0.555 mSv.

1. INTRODUCTION

Radionuclides are naturally present in the environment and in all living things, food and water. All beings are exposed to natural radiation (also called background radiation) on a day to day basis [1]. The radiation originates from radionuclides of natural origin found in soil, water and air. Radioactivity can be detected in food and water, and the radionuclide concentration varies according to several factors such as local geology, climate and agricultural practices [2]. Radioactive material is transferred into the food chain in the same way as non-radioactive material. The degree of harm to human health depends on the type of radionuclide and the period of exposure [1]. High background radiation areas (HBRAs) have attracted much interest from the point of view of radiological protection. Studies that evaluate the biological harm in humans exposed to radionuclides in the ^{238}U and ^{232}Th series, which have long half-lives, are very important [3]. Among the HBRAs, the Poços de Caldas plateau, located in south-eastern Brazil, is known worldwide for exhibiting several radioactive anomalies. The situation of the population residing in the cities affected by the volcanic region of Poços de Caldas is of special interest for public health surveillance related to physical factors, presenting a rich framework for studies that improve the knowledge about the long term exposure to natural radioactivity in regions with geological anomalies with high concentrations of radioactive minerals. The World Health Organization recommends the 'total diet study' (EDT) as the most appropriate method for estimating the intake of contaminants and nutrients for a country or large population groups [1] .The use of the 'family budget researches' (POFs) of the Brazilian Institute of Geography and Statistics (IBGE) is one of the ways to determine the food consumption data for use in EDTs. The objective of this work is to determine which of the radioactive elements ^{40}K, ^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{232}Th, ^{230}Th, ^{228}Th, ^{234}U, ^{235}U and ^{238}U are radiologically relevant and present in the diet of the population in the HBRA of south-eastern Brazil.

2. METHODS

The sampling and preparation of food were carried out using the groups described in the 2008–2009 IBGE POF [4], and each group was regarded as a composite, with a total of 83 types of food divided into 20 groups. The composites were prepared according to the food as they are consumed. The food eaten after cooking was prepared without the use of spices, condiments, oils and fats, and other ingredients were not introduced during the preparation, except for the addition of distilled water [5]. After preparation, each composite was homogenized manually or using a home processor, then the composites were dried at 75°C, crushed, dissolved with nitric acid and perchloric acid, and forwarded for radiometric analysis.

2.1. Gamma spectrometry

One of the advantages of gamma spectrometry is the ability to determine radionuclides without the need for chemical separation and acid dissolution. The equipment has a semiconductor detector of high purity germanium (HPGe), which has a good energy resolution and is connected to multi-channel analysers and the appropriate software for identification and quantification of radionuclides. This methodology was used for the determination of ^{40}K.

2.2. Alpha spectrometry

This method is used to determine the alpha radiation emitting isotopes previously isolated from the matrix and purified by radiochemical separation. The source for the measurement of isotopes is extremely thin, and in this work it was obtained by the methods of electroplating and spontaneous deposition. This technique was used in this work for the determination of ^{210}Po, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U.

2.3. Alpha and beta total counting

This method is used for the determination of the radionuclides ^{210}Pb, ^{226}Ra and ^{228}Ra, and is based on chemical separation from the other elements present in the sample, using a purification technique with selective co-precipitation. It is an indirect method of determination based on the short half-life progeny of the radionuclides to be quantified. This technique has an advantage over gamma spectrometry, as it presents a low natural radiation and consequently has also a very low detection limit for these same radionuclides.

3. RESULTS AND DISCUSSION

The reference material IAEA-327-Soil was analysed in order to validate the methodologies used for the radionuclide determinations and quality control of the results. The matrix used for the validation was a soil, which could be considered a more complex matrix than food. The activity concentrations of ^{40}K, ^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U showed that the methods used to determine these radioisotopes are reliable and accurate. The doses received by food intake by population groups can be calculated as follows:

$$E_A = \sum_j \sum_f C_{j,f} \times M_{j,A} \times T \times h_{j,A} \qquad (1)$$

where:

E_A is the effective dose absorbed per group (mSv),

$C_{f,j}$ is the average radionuclide concentration in the food (Bq/kg),

$M_{j,A}$ is the mass consumed by group A (kg/d),

T is the total length of time considered (d).

$h_{j,A}$ is the ingestion dose coefficient for the radionuclide for Group A (mSv/Bq).

The risk associated with the ingestion of radionuclides by humans is proportional to the total ingested dose of radionuclides. In general, it is assumed that the stochastic effects increase linearly with dose and the effective dose is usually used to define such risk. Table 1 presents the total contribution of each radionuclide to the total annual effective dose.

TABLE 1. EFFECTIVE DOSE

	Dose coefficient (mSv/Bq)	Effective dose (mSv/d)
K-40	6.20×10^{-6}	7.53×10^{-4}
Pb-210	6.80×10^{-4}	4.18×10^{-4}
Po-210	2.40×10^{-4}	6.26×10^{-5}
Ra-226	2.80×10^{-4}	4.41×10^{-5}
Ra-228	6.70×10^{-4}	2.24×10^{-4}
Th-228	7.20×10^{-5}	1.54×10^{-5}
Th-230	2.10×10^{-4}	9.99×10^{-7}
Th-232	2.20×10^{-4}	9.79×10^{-7}
U-234	4.90×10^{-5}	8.28×10^{-7}
U-235	4.60×10^{-5}	2.63×10^{-8}
U-238	4.40×10^{-5}	5.42×10^{-7}
Total		1.52×10^{-3}
Total effective dose in a year (mSv)		0.555

Note: The dose coefficients are those for ingestion by adults, as published in 2012 by the International Commission on Radiological Protection.

In order to verify the radiological dietary safety of the population of an HRBA in Nigeria (Bitsichi, Jos Plateau), activity concentrations of ^{226}Ra, ^{228}Th and ^{40}K in food samples and soil of the region were analysed. The dose rates that were found ranged from 0.50 to 1.47 μSv/h. However, the values obtained suggested that the dose of these radionuclides in food intake was low and that detrimental health effects were not expected [6]. The values found for this Nigerian study were much higher than the total annual effective dose determined in the present work (0.555 mSv/a, implying a dose rate of 0.06 μSv/h), indicating that doses received by ingestion are not harmful to the health of the Poços de Caldas Plateau population. The measurement of radioactivity in the total diet food groups is extremely important for monitoring the radiation levels to which humans can be directly or indirectly exposed, in particular the determination of radionuclides of the ^{238}U and ^{232}Th series, as presented to the population of Pocos de Caldas, a city with high natural radioactivity.

4. CONCLUSION

The total annual effective ingestion dose received by the urban population on the Pocos de Caldas plateau was determined to be 0.555 mSv. This does not give rise to any health concern.

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Poster Presentation

RADIONUCLIDES OF NATURAL ORIGIN IN A BRAZILIAN MUSSEL REFERENCE MATERIAL

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Abstract

Certified reference materials are important tools for the quality assurance of analytical results. However there are several constraints for their widespread use in developing countries such as lack of technological development or difficult access to imported goods. Another issue is that analyte level differences between imported certified reference materials and local laboratory samples may be a concern in the measurement process. This contribution presents the activity concentrations of ^{234}U, ^{235}U, ^{238}U and ^{232}Th determined by alpha spectrometry after a radiochemical separation procedure and ^{40}K and ^{210}Pb determined by gamma-ray spectrometry in a *Perna perna* mussel reference material produced in Brazil. The activity concentrations obtained were included as information values in the certification process of the reference material.

1. INTRODUCTION

Certified reference materials are used for quality assurance of analytical results, method validation, quality control and to establish the metrological traceability of measurement results [1]. Their production is a complex task involving several steps such as preliminary tests, sampling, sample preparation, particle size and humidity adjustment, property characterization and certification [2]. Due care in the production process is essential in order to guarantee that the properties of interest are well established and that certified values are accompanied by appropriate expanded uncertainties that take into consideration not only the characterization process but also the stability and the homogeneity of the material [2].

Biological reference materials prepared from animal tissues such as mussels and oysters are useful in the quality assurance of environmental and nutritional studies. These materials usually present certified values for chemical elements and species, organic pollutants, toxins and microorganisms. However, few biological reference materials are available for naturally occuring and/or artificial radionuclides such as IAEA-437 mussel and IAEA-414 fish flesh reference materials [3–5].

Uranium and thorium series radionuclides are ubiquous in the marine environment. They may originate from natural processes as well as from anthropogenic activities such as mining industries and disposal of radium rich products such as phosphogypsum [6]. These radionuclides are prone to bioaccumulation in marine organisms and have implications for environmental biomonitoring studies [7]. Observed concentrations of the radionuclides ^{210}Po, ^{210}Pb or ^{232}Th in the biota are subject to environmental conditions as the salinity or temperature of marine water, season of the year and to biological parameters such as size (shell length and soft tissue weight) or sexual maturation, which complicates result interpretation [7, 8].

From a human health perspective, the determination of naturally occuring radionuclides and associated activity concentrations, particularly for alpha emitters, is important for the estimation of the intake levels from the consumption of marine foodstuffs such as mussels or oysters [9, 10].

This study presents the activity concentrations of ^{234}U, ^{235}U, ^{238}U, ^{232}Th, ^{210}Pb and ^{40}K in a *Perna perna* mussel reference material produced in Brazil as a contribution to the quality assurance of radionuclide measurements performed in the country.

2. METHODS

2.1. Mussel reference material production

For the preparation of the Brazilian mussel reference material, 164 kg of *Perna perna* (Linnaeus, 1758) mussels were purchased from a single producer, from Cocanha Beach in Caraguatatuba City, São Paulo State north shore. Soft tissues were homogenized in a domestic blender adapted with titanium blades. This process yielded approximately 36 kg of material which was freeze dried in a Thermo Savant Modulyo D freeze dryer, resulting in 5.4 kg of freeze dried material. The material was ground in the blender and the portion with particle size above 105 µm was discarded. The powder was homogenized for 72 h in a Y type homogenizer. Then the bulk material was bottled in 171 bottles containing approximately 13 g each. Afterwards the material was irradiated with a gamma dose of 5 kGy to enhance its stability [11].

2.2. Alpha spectrometry

The alpha spectrometry methods used are described in detail elsewhere [12–14]. Subsamples of approximately 5 g were burnt to ash at 450°C for 24 h in a muffle furnace. Then the ash samples were dissolved with three acids (nitric, perchloric, and hydrofluoric). The solution was evaporated and reconstituted with 8 mol/L nitric acid.

After sample digestion, ^{229}Th and ^{232}U radiotracers were added in order to determine the chemical recuperation. Thorium and uranium radionuclides were separated and purified using specific ion exchange resins (DOWEX 1x2 and UTEVA, respectively) for sequential chromatography extractions and were electrodeposited on polished silver plates. An Alpha Analyst spectrometer with 12 passivated implanted planar silicon (PIPS) detectors (counting efficiency 18%), and Genie 2000/Alpha Analyst spectroscopy systems, from Canberra Industries was used for radionuclide quantification. Samples were measured for 200 000 s. Alpha particle energies of 4.90 MeV for the ^{229}Th tracer and 4.01 MeV for ^{232}Th were used to quantify thorium. Alpha particle energies of 4.31 MeV for the ^{232}U tracer, 4.74 MeV for ^{234}U, 4.47 MeV for ^{235}U, and 4.19 MeV for ^{238}U were used to quantify uranium.

For expanded uncertanties estimation, the GUM software was employed using parameters associated with alpha spectrometry and the main uncertainty sources were identified for this radioanalytical method. Most sources of uncertainty in radioanalytical measurements, such as the sources in the intercomparison exercise, were classified as a normal probability distribution. The uncertainty of the detector efficiency was estimated from a series of repeated observations by calculating the standard deviation of the mean (approximately 20 experimental measurements for uranium and 20 experimental measurements for thorium) [15, 16].

2.3. Gamma-ray spectrometry

Reference material subsamples of approximately 30 g were kept in sealed plastic vials. A Canberra model GX4510 high purity germanium (HPGe) detector was used for ^{40}K detection via the 1461 keV photopeak and ^{210}Pb was detected using the X ray characteristic photopeak at 46.5 keV after a self-absorption correction. Genie 2000 software was used for data acquisition and treatment. The spectrometer was calibrated in energy using ^{152}Eu and ^{210}Pb certified sources (IRD/CNEN-RJ) and the calibration in efficiency was performed using standard solutions of the same radionuclides added to alumina Suprapur, kept in a vial with the same geometry as that of the samples. The average counting period was 250 000 s.

3. RESULTS AND DISCUSSION

Table 1 presents activity concentrations obtained for the mussel reference material on a dry mass basis. As results were reported by only one laboratory, the values were considered as informative rather than certified values. It was observed that while the activity concentrations of uranium series radionuclides were of the same order of magnitude as the ones reported for IAEA-437 and IAEA-414 reference materials, ^{40}K and ^{210}Pb presented much lower activity concentrations and ^{232}Th presented activity concentrations approximately 10 times higher if compared with IAEA-437 mussel reference material.

TABLE 1 INFORMATIVE VALUES OF ACTIVITY CONCENTRATION FOR THE MUSSEL REFERENCE MATERIAL (DRY MASS BASIS).

Radionuclide	Activity concentration, Bq/kg
^{40}K	0.0446 ± 0.0031
^{210}Pb	0.106 ± 0.032
^{232}Th	1.59 ± 0.62
^{234}U	1.39 ± 0.52
^{235}U	0.067 ± 0.065
^{238}U	1.25 ± 0.54

Note: Reported uncertainties are expanded uncertainties with a coverage factor $k = 2$ which gives a level of confidence of approximately 95%. Reference date for decay correction: 23 April 2009.

4. CONCLUSIONS

This study presents the informative values for the activity concentration of ^{234}U, ^{235}U, ^{238}U and ^{232}Th determined by alpha spectrometry after a radiochemical separation procedure and activity concentrations of ^{40}K and ^{210}Pb determined by gamma ray spectrometry in a *Perna perna* mussel reference material produced in Brazil. The reference material is intended to be used as a contribution to the quality assurance of radionuclide measurements performed in biological samples of marine origin in the country.

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Poster Presentation

NATURAL RADIOACTIVITY IN BRAZILIAN UNDERGROUND MINES

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Abstract

Radionuclides of natural, terrestrial origin are present in many natural resources. The radionuclides of interest are those in the ^{238}U, ^{232}Th decay chains, including radon and its short-lived progeny, and ^{40}K. The activity concentrations of these radionuclides depend on the type of mineral and its origin. Owing to local geological and geochemical factors, some areas have elevated levels of terrestrial radiation and are commonly referred to as high background radiation areas. Industrial activities involving the processing of minerals can give rise to exposure to material with elevated concentrations of radionuclides, some of which may need some form of regulatory control and may therefore fall within the definition of NORM. This work presents the results of a characterization of radioactivity levels in Brazilian underground mines. Radon concentrations were measured using AlphaGUARD detectors. The radon progeny concentrations were determined using a DOSEman detector. The equilibrium factors between radon and its short-lived progeny were also determined. In situ measurements of external gamma dose rate at 1 m from the rock face were performed. Based on these data, the internal and external annual effective doses received by mine workers were estimated. In addition, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in rock and soil samples were determined by means of gamma ray spectrometry using a high purity germanium detector, while radon concentrations in groundwater samples were measured using a RAD7 alpha spectrometric detector. Radon gas concentrations ranged from 60 to 8171 Bq/m^3 while radon progeny equilibrium equivalent concentrations ranged from 12 to 1174 Bq/m^3. The measurement results and the estimated effective doses will be used as an input to the development of Brazilian regulatory standards.

1. INTRODUCTION

Humans and the environment are exposed to ionizing radiation from natural and artificial sources. Natural radioactivity is the largest contributor to the dose received by members of the public. Apart from cosmic rays, the main sources of natural radioactivity are the radionuclides of natural origin that are widely distributed throughout the environment. Such radionuclides are classified into three categories: cosmogenic, primordial and secondary radionuclides [1]. In terms of radiological impact, the radionuclides of interest are those in the ^{238}U and ^{232}Th decay series, including radon and its short-lived progeny (secondary radionuclides) and, to a much lesser extent, the primordial radionuclide ^{40}K [2]. The activity concentrations of these radionuclides in the environment depend on the type of mineral and its origin [3]. Owing to local geological and geochemical factors, some areas have elevated levels of terrestrial radiation and are commonly known as high background radiation areas [4]. Igneous rocks such as granite commonly have elevated radionuclide concentrations. More moderate activity concentrations are generally observed in sedimentary rocks, although there are exceptions to this rule such as phosphate rich rock and some types of shale [1]. Industrial activities involving the processing of minerals can give rise to exposure to material with elevated concentrations of radionuclides, some of which may need some form of regulatory control and may therefore fall within the definition of NORM. During mining operations, mine workers receive internal exposure to radon and its short-lived progeny through inhalation, as well as external exposure to gamma radiation. Radon-222 decays by emitting alpha particles, generating a sequence of progeny (isotopes of the metals Po, Pb and Bi) in the form of positively charged atoms. Some of these attach to the surrounding aerosol particles in the air, forming 'attached radon progeny'. The radon progeny fraction that does not attach to aerosol particles in the air ('unattached radon progeny') may become deposited on surfaces or may continue to exist as free ions or small molecular clusters [5, 6]. When inhaled, both the attached and unattached radon progeny may deposit in the respiratory system, especially in the upper respiratory tract, irradiating lung tissue [6]. The International Agency for Research on Cancer classifies radon as a class I carcinogen [7] .Short-lived radon progeny are recognized as the main radiological health risk in underground mines [8].

The objective of the present study was to characterize the radiological conditions in underground mines and to assess the implications in terms of possible health hazards to workers. The work is a part of a radon project that is being conducted by the National Commission on Nuclear Energy. The National Department of Mineral Production reports on the existence of underground mines in Brazil. Many of these mines have uranium and thorium mineralization [9]. The data generated in this study will contribute to the control of occupational exposure and to the preparation of Brazilian regulatory standards.

2. MATERIALS AND METHODS

Activity concentrations of ^{222}Rn and progeny, ^{226}Ra, ^{232}Th and ^{40}K were determined in eight underground mines in which agalmatolite, coal, emeralds, fluorite, scheelite, tourmaline and zinc were being exploited. Drilling, blasting, materials transport and mine maintenance are examples of the typical activities in these mines. The radon concentration measurements were carried out with an AlphaGUARD (Saphymo model PQ2000PRO) operating as a pulse ionization chamber. The monitor was programmed in the diffusion mode at intervals of 10 or 60 min, acting as a continuous passive detector. Measurements of the short-lived radon progeny (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) were performed by alpha spectrometry using a solid state alpha detector (Sarad DOSEman PRO) to determine the potential alpha energy concentration and equilibrium equivalent concentration (EEC). The DOSEman detector was programmed at intervals of 60 min. The AlphaGUARD and DOSEman were installed together in each of the underground mines at a location corresponding to the maximum occupancy time, and remained protected and secure for approximately 2 d. The arithmetic mean radon and radon progeny concentrations were obtained for each mine. In accordance with Refs [1, 10], the annual effective dose from inhalation of radon progeny was calculated from Eq. (1).

$$E = C_{Rn} \times F \times T \times k \tag{1}$$

where E is the annual effective dose (mSv/a),
C_{Rn} is the radon concentration (Bq/m^3),
F is the equilibrium factor (the EEC divided by the radon concentration,
T is the occupancy time (2000 h/a),
k is the dose conversion factor (9×10^{-6} mSv per Bq·h/m^3).

The mean annual gamma dose at each mine was determined by direct measurement of the gamma dose rate (in μSv/h) using an Identifinder (THERMO) and RS-230 (Radiation Solutions Inc.) at points distributed throughout the mine workings. The mean of the dose rate measurements was converted to annual effective dose (*D*) by assuming an occupancy time of 2000 h [10]. The total annual effective dose was therefore the sum of *E* and *D*.

Concentrations of radon in the underground water were measured using a RAD7 (Durridge Company Inc.) alpha spectrometric detector. The water was collected directly from the rock fissures without contact with air. The analytical technique used for determining the activity concentrations of ^{226}Ra (in the ^{238}U decay series),^{232}Th and ^{40}K was gamma spectrometry using a Canberra high purity germanium detector having 15% efficiency and coaxial geometry in a very low background counting room. Samples of various geological materials from each mine were collected, powdered, sieved and transferred to 1 L Marinelli beakers. Each beaker remained tightly sealed for 30 d in order to restore secular equilibrium between ^{226}Ra and its progeny. The ventilation conditions within each mine were determined using an anemometer (Instrutherm AD-250) at the same locations as those of the gamma dose rate measurements.

3. RESULTS AND DISCUSSION

Preliminary results of this study were reported in Refs [11, 12]. The air velocity, the concentrations of radon and its progeny in underground air (determined by continuous measurement), the equilibrium factor *F* and the estimated total effective dose received by workers are shown in Table 1 for each mine. The air velocity ranged from <0.1 to 1.8 m/s in operating mines. In two instances, the mine was not in operation and the ventilation system had been turned off.

TABLE 1. AIRBORNE RADON AND RADON PROGENY, AND GAMMA RADIATION

Mine	Air velocity (m/s)	Activity concentration in air (Bq/m^3)				Equilibrium factor	Effective dose (mSv/a)		
		Radon		Radon progeny (EEC)			Radon progeny	Gamma radiation	Total
		Mean	Range	Mean	Range				
A[a]	–[a]	3889	528–4128	1174	43–1317	0.3	21	1	22
A	0.7	714	299–2088	377	42–1581	0.5	7	1	8
B	1.8	949	584–1264	259	123–444	0.3	5	2	7
C	<0.1	113	38–192	76	26–144	0.7	1	0.3	1
D[a]	–[a]	4964	1392–10880	1148	209–2765	0.2	21	0.2	21
E	<0.1	1442	792–2288	228	96–357	0.2	4	0.4	5
F	–[b]	327	65–617	141	19–279	0.4	3	0.8	4
G	<0.1	326	272–380	213	179–249	0.7	4	0.1	4
H	<0.1	60	39–99	12	3–36	0.2	0.2	0.3	1

[a] Not in operation.
[b] Not measured.

The concentrations of radon and its progeny varied considerably from mine to mine and within the same mine. This was a consequence of certain characteristic factors in each mine, including the ^{226}Ra activity concentration in the rock, the permeability and porosity of the rock, the ventilation conditions, the temperature, the humidity and the atmospheric pressure. According to Ref. [3], the most important factor is ventilation, which varies between and within mines. At certain times during the measurement period, the radon concentrations in mines A, B, D and E exceeded the proposed reference level of 1000 Bq/m^3 [13, 14]. The mean radon progeny concentration ranged from 12 to 1174 Bq/m^3. The equilibrium factor depends on the ventilation rate and age of the air at a particular location, and on the radon exhalation and dilution rate — the wide range of equilibrium factors observed in this study (0.2–0.7) suggests that the adoption of a mean equilibrium factor for dose assessment could lead to significant errors.

The main contribution to the total annual effective dose was that from radon progeny. A radon concentration at a reference level of 1000 Bq/m^3, assuming an equilibrium factor of 0.4, would lead to an annual effective dose (E) of 7 mSv/a [13]. As shown in Table 1, in two mines (A and D) the value of E was above this level of effective dose and the total annual effective dose received by underground workers would have exceeded the occupational exposure limit of 20 mSv/a [15]. However, the calculation of dose in these instances was not really meaningful because the two mines were not in operation at the time and the ventilation systems had been turned off. For the mines in operation, the total annual effective dose received by underground workers due to exposure to radon progeny (E) and gamma radiation (D) ranged from 1 to 8 mSv/a.

Table 2 shows the radon concentrations in underground water and the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in rock and soil samples collected from the surveyed mines. For comparison, the worldwide average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil are 32, 45 and 420 Bq/kg, respectively [1].

TABLE 2. RADIONUCLIDES IN UNDERGROUND WATER AND IN ROCK AND SOIL

Mine	Radon activity concentration in underground water samples (Bq/L)			Activity concentration in rock and soil samples (Bq/kg)						
				Number of samples	Ra-226		Th-232		K-40	
	Number of samples	Mean	Range		Mean	Range	Mean	Range	Mean	Range
A	4	487	336–637	4	75	41–120	131	67–187	1278	604–1827
B	2	851	786–915	8	339	8–517	19	6–51	227	10–616
C	2	25	24–26	6	39	1–182	17	1–43	331	6–1146
D	3	6	3–8	6	64	11–297	16	0.4–39	563	57–2852
E	3	8	6–9	6	21	8–60	8	3–27	693	1–2371
F	3	13	11–15	5	28	11–71	36	3–90	718	162–1014
G	3	2	2–4	4	5	3–8	5	3–6	187	36–408
H	No underground water			4	63	13–145	16	2–39	201	16–665

In mine A, all samples collected had elevated radionuclide activity concentrations. In mine B, all samples had an elevated ^{226}Ra activity concentration, but the ^{232}Th and ^{40}K activity concentrations were close to background levels. These mines had higher radon concentrations in the underground water. This correlated with the elevated radon concentrations found in those mines (Table 1). In mines C, D, E and H, only the pegmatite samples of different origin exhibited a ^{226}Ra activity above the average value for soil worldwide. The ^{232}Th activity concentrations were very low, while the ^{40}K activity concentration was elevated in some samples. In mine F, only the soil samples had elevated ^{226}Ra and ^{232}Th activity concentrations, not the samples of rock collected from within the mine. Four samples from mine F exhibited elevated ^{40}K activity. In mine G, the only significant activity concentration was that of ^{40}K. Mines C, D, E, F and G also did not have high radon concentrations in the underground water. Therefore, the locations with enhanced radon concentration in mines D and E may be a consequence of radon exhalation from pegmatite. Mines C, F, G and H had radon concentrations below those in the other mines. A possible explanation is that mines C and F are smaller than the others and thus have shorter ventilation pathways and consequently better ventilation, while mines G and H had large, high workings, with communication to the surface in certain places. Nevertheless, mines C and H had pegmatite containing significant concentrations of ^{226}Ra.

4. CONCLUSIONS

Radon concentrations vary considerably from mine to mine and within the same mine. Radon concentrations above the proposed reference level of 1000Bq/m^3 were observed in mines A, B, D and E, implying that remedial action might be justified, for example by improving the ventilation or modifying work patterns to reduce the exposure time. Equilibrium factors varied from mine to mine, resulting in a mean value of 0.4, but the wide range of values (0.2–0.7) suggests that using this mean value for the assessment of dose could lead to significant errors. Elevated ^{226}Ra activity concentrations were found in all mines except mines E, F and G. Only mines A and B exhibited ^{232}Th activity concentrations above the worldwide average for soil. Potassium-40 activity concentrations above or near the reference level were found in at least one sample from each mine. For mines in operation, the total annual effective dose estimated for workers varied from 1 to 8 mSv/a. The results suggest that measurements of radon and its progeny should be conducted periodically in Brazilian underground mines.

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Poster Presentation

RADIONUCLIDES OF NATURAL ORIGIN IN ENVIRONMENTAL SAMPLES COLLECTED IN THE CATCHMENT AREA OF PONTE NOVA RESERVOIR, SÃO PAULO, BRAZIL

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Abstract

Natural radiation is generally classified as terrestrial primordial radiation or cosmogenic radiation. Primordial radiation is mostly due to the decay series of ^{238}U and ^{232}Th and is present in soil, sediments and water; another important source of natural radiation is ^{40}K. Reservoirs are a very dynamic system with several phenomena to study, due to their huge impact on ecosystems and river flow. Ponte Nova reservoir (23°34'43.23"S, 45°56'56.76"W) is the first reservoir in a cascade system that was built in the 1970s to control the Upper Tietê River basin water flow. In the present work, the activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Th, ^{228}Ra and ^{40}K were determined using instrumental neutron activation analysis and gamma spectrometry in soil profiles collected in the catchment area of Ponte Nova Reservoir and sediment cores collected close to the soil samples. The highest concentration obtained in the soil samples was for ^{40}K that varied from 49 to 2410 Bq/kg and ^{210}Pb in the sediment samples, which varied from 20 to 774 Bq/kg. Cluster analysis and principal component analysis were applied to all the results obtained to verify a probable correlation between the radionuclides determined in the soil and sediment samples.

1. INTRODUCTION

Natural radioactivity characterization is the basis of radioecology and environmental monitoring [1]. The average worldwide dose due to natural radiation is 2.4 mSv/a [2], although individual values can be considerably higher. Natural radiation can be classified as terrestrial primordial radiation or cosmogenic radiation. Primordial radiation is mostly due to the decay series of ^{238}U and ^{232}Th and is ubiquitous in soil, sediments, and water. Another important source of natural radiation is ^{40}K, which is present in a wide range of rocks and soil [3]. These rocks underwent and still undergo weathering processes which in geological time turns them into the soil; eventually, this soil will be carried by wind and water and will become part of rivers, sediments, lakes, reservoirs, and oceans, as well as absorbed by fauna and flora [4]. Although slower moving than rivers, reservoirs are very dynamic systems with a large range of phenomena to study, due to their huge impact on ecosystems and river flow. As a result of a drought in the south-eastern region of Brazil in the years 2013–2015 [5], the Upper Tietê basin was severely affected and focused the attention of the media and the general population on Brazilian water resources. The Ponte Nova reservoir, located in the state of São Paulo, is the first reservoir in a cascade system that was built in the 1970s to control the Upper Tietê River basin water flow. This water basin serves millions of people with water in the São Paulo metropolitan area and the São Paulo greenbelt that provides this metropolitan area with agricultural products [6]. In the present work, the activity concentrations of ^{238}U and ^{232}Th were determined by instrumental neutron activation analysis (INAA) and ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th and ^{40}K by gamma spectrometry in soil profiles collected in the catchment area of Ponte Nova reservoir, and also in sediment cores collected close to the soil samples.

2. METHODOLOGY

Soil samples were collected vertically at three locations in the main lithology of the reservoir, from the top down to 1 m, sampled every 5 cm. The sediment cores were sliced every 3 cm. The real and apparent soil density and grain size analysis were determined in accordance with the manual of the Brazilian Agricultural Research Corporation [7]. Grain size analysis was performed on the soil and sediment samples by sieving 5 g of each sample with Milli-Q water in sieves capable of separating the soil fractions in sand, silt and clay and the sediment fractions in sand and silt–clay. The activity concentrations of ^{238}U and ^{232}Th were determined by INAA. Approximately 200 mg of the soil and sediment samples and IAEA reference material SL1 and Montana II (NIST) were irradiated for 6 h in a thermal neutron flux of 1012 n·cm^{-2}·s^{-1} at the IEA-R1 Research Reactor at Instituto de Pesquisas Energéticas e Nucleares [8]. The activity concentrations of ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th and ^{40}K were determined by gamma-ray spectrometry. Approximately 100 g of the soil and 15 g of the sediment samples were measured in a coaxial Be-layer high purity germanium detector with 25% relative efficiency, 2.09 keV resolution at 1.33 MeV and associated electronic devices, with a live counting time of 150 000 s. The spectra were acquired by an Ethernim multi-channel analyser and, for the analysis, WinnerGamma software was used [8].

3. RESULTS AND DISCUSSION

The grain size analysis showed that all the soil and sediment samples were composed predominantly of sand, with some exceptions in both matrixes, classifying them as sandy clay (see Figs 1 and 2). The soil samples presented a mean apparent density of 1.11 g/cm^3 and a real density of 2.96 g/cm^3, which implies a mean porosity of 56.57%, indicating a high permeability of the soil, allowing the radionuclides to move easily through the layers of soil.

Table 1 shows the activity concentrations measured in the soil and sediment samples. Soil profile 1 shows a higher activity concentration for ^{40}K and ^{232}Th than the other profiles. Soil profile 2 shows a similar activity concentration for ^{238}U and ^{232}Th, and soil profile 3 shows the lowest concentration of ^{232}Th. These differences may be due to differences in lithology.

Sediment cores 1 and 2 showed a higher concentration of ^{210}Pb than sediment core 3, which is located upstream and close to the source of the Tiête River and may be less affected by human activity. In cores 1 and 2 the ^{210}Pb activity concentration was elevated in relation to ^{226}Ra — the two locations are close to a highway and several farms. The mean worldwide values for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in soil are 33, 32, 45 and 412 Bq/kg, respectively [2]. Comparing the values obtained in this work with the worldwide values shows that the majority of the samples exhibited elevated values.

FIG. 1. Grain size results for soil.

FIG. 2. Grain size results for sediment.

TABLE 1. ACTIVITY CONCENTRATION RESULTS FOR SOIL
(Uncertainties in parentheses)

Depth (cm)	Activity concentration (Bq/kg)						
	U-238	Ra-226	Pb-210	Th-232	Th-228	Ra-228	K-40
Soil profile 1:							
0–5	41(4)	37(2)	95(10)	71(4)	126(13)	84(6)	639(94)
5–10	46(3)	41(2)	88(9)	99(7)	130(12)	102(6)	649(45)
10–15	42(4)	43(2)	80(7)	155(8)	177(17)	106(6)	780(52)
15–20	49(4)	47(2)	43(6)	111(8)	168(15)	139(8)	1157(79)
20–25	53(4)	40(2)	44(6)	128(9)	179(17)	128(8)	1208(82)
25–30	43(3)	31(1)	31(6)	117(8)	175(23)	143(9)	1358(131)
30–35	42(5)	38(2)	45(5)	207(10)	155(14)	140(8)	1430(96)
35–40	40(3)	40(1)	41(5)	107(8)	161(16)	127(8)	601(41)
40–45	49(3)	39(1)	49(6)	166(8)	137(14)	113(7)	636(44)
45–50	33(3)	37(2)	48(6)	90(6)	123(12)	99(6)	510(36)
50–55	35(3)	37(2)	44(5)	142(7)	111(11)	99(6)	312(22)
55–60	42(2)	33(2)	40(5)	124(6)	116(11)	87(5)	178(13)
60–65	43(2)	33(2)	42(5)	116(6)	108(11)	92(6)	169(13)
65–70	34(2)	31(1)	37(3)	123(6)	109(15)	89(6)	178(18)
75–80	34(3)	32(1)	35(5)	147(7)	143(14)	93(6)	155(23)
80–85	41(2)	34(2)	22(3)	–	133(13)	104(6)	167(12)
85–90	49(4)	36(1)	35(7)	118(6)	102(14)	113(7)	144(16)
90–95	62(4)	33(1)	30(4)	165(8)	59(6)	66(4)	129(19)
95–100	34(3)	33(1)	39(6)	66(3)	32(6)	47(3)	161(17)
Range	*33–62*	*31–47*	*31–95*	*71–207*	*32–179*	*47–143*	*129–1430*
Soil profile 2:							
0–5	32(2)	25(2)	45(5)	33(2)	31(3)	26(2)	49(8)
5–10	44(2)	30(3)	50(6)	51(3)	36(5)	33(2)	59(9)
10–15	44(2)	38(2)	51(6)	48(2)	46(3)	37(3)	69(18)
15–20	53(3)	40(4)	50(6)	54(3)	46(3)	40(3)	64(10)
20–25	49(3)	33(1)	23(3)	51(3)	37(2)	36(2)	60(5)
35–40	52(3)	41(4)	39(4)	59(3)	52(3)	47(3)	64(10)
55–60	53(3)	42(2)	47(5)	55(3)	63(3)	50(3)	65(5)
65–70	56(3)	35(1)	26(3)	69(3)	47(2)	66(4)	64(5)
80–85	42(3)	32(1)	25(3)	55(3)	41(2)	42(2)	63(5)
100–105	53(3)	45(2)	40(5)	59(3)	62(3)	56(4)	113(17)
110–115	40(2)	38(2)	29(3)	50(3)	56(3)	51(3)	88(6)
120–125	48(3)	45(2)	46(5)	56(3)	64(3)	53(3)	101(8)
Range	*32–56*	*25–45*	*23–51*	*33–69*	*31–64*	*26–66*	*49–113*
Soil profile 3:							
0–5	36(4)	38(3)	39(5)	11(1)	46(6)	37(3)	69(11)
10–15	50(4)	44(2)	33(4)	13(1)	61(3)	53(4)	61(10)
20–25	66(6)	48(2)	21(2)	17(1)	59(3)	58(3)	57(5)
30–35	70(6)	47(2)	21(2)	19(1)	63(3)	60(3)	55(4)
40–45	54(5)	46(2)	28(3)	16(1)	73(3)	56(3)	44(4)
50–55	63(5)	46(2)	33(3)	17(1)	78(4)	61(4)	55(4)
Range	*36–70*	*38–48*	*21–39*	*11–19*	*46–78*	*37–61*	*44–69*

TABLE 2. ACTIVITY CONCENTRATION RESULTS FOR SEDIMENT
(Uncertainties in parentheses)

Depth	Activity concentration (Bq/kg)						
(cm)	U-238	Ra-226	Pb-210	Th-232	Th-228	Ra-228	K-40
Sediment core 1:							
0–6	65(4)	58(8)	105(24)	203(10)	181(12)	175(15)	–
6–9	73(4)	74(4)	356(52)	247(12)	157(15)	174(15)	–
9–12	65(4)	75(6)	306(49)	372(18)	319(20)	294(25)	–
15–18	42(3)	36(3)	178(35)	124(6)	101(8)	93(8)	–
21–24	20(1)	22(2)	108(26)	36(2)	22(2)	24(2)	–
27–30	49(3)	23(2)	75(26)	83(1)	26(4)	21(2)	–
33–36	38(2)	27(2)	133(17)	23(1)	24(2)	32(3)	–
Range	*20–73*	*22–75*	*75–356*	*23–372*	*22–319*	*21–294*	
Sediment core 2:							
0–5	89(5)	82(7)	774(99)	96(5)	76(15)	68(7)	–
14–17	63(4)	24(3)	132(28)	42(2)	21(8)	26(3)	–
32–35	68(3)	76(6)	401(56)	60(3)	43(11)	54(5)	–
41–44	68(3)	62(5)	252(40)	53(3)	41(10)	40(4)	–
44–47	16(1)	72(4)	308(46)	12(1)	44(12)	50(5)	–
53–56	74(3)	62(4)	212(36)	62(3)	43(10)	45(4)	–
56–59	67(3)	70(4)	266(42)	62(3)	47(12)	52(5)	–
62–65	69(4)	48(3)	186(34)	68(3)	36(9)	34(3)	–
68–72	86(5)	60(4)	249(40)	71(4)	48(10)	51(5)	–
Range	*16–89*	*24–82*	*132–774*	*12–96*	*21–76*	*26–68*	–
Sediment core 3:							
0–5	50(4)	38(2)	47(7)	65(3)	62(5)	41(4)	148(14)
8–11	50(5)	54(3)	63(8)	71(3)	76(5)	62(5)	168(14)
14–17	54(5)	68(3)	33(4)	80(4)	70(4)	71(5)	–
20–23	76(6)	75(4)	37(5)	94(5)	78(6)	81(6)	–
26–29	66(6)	66(4)	69(8)	98(5)	79(5)	64(6)	154(14)
Range	*50–76*	*38–75*	*33–69*	*65–98*	*62–79*	*41–81*	*148–168*

4. CONCLUSION

Soil and sediment samples showed an overall association with the lithology and the grain size of the samples. Radionuclide activity concentrations were consistent with average and elevated values in soil reported worldwide.

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Poster Presentation

EXPERIENCE AND PERFORMANCE OBTAINED IN INDOOR RADON INTERCOMPARISON EXERCISES

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Abstract

Awareness of the effects of human exposure to radon grows as studies are conducted worldwide. Today, intercomparisons are useful tools in the search for measurement improvements and reliable results. As a part of the National Commission on Nuclear Energy, the Poços de Caldas Laboratory performs indoor radon measurement studies aimed at the validation of a solid stated nuclear track detector technique, working with passive detector analyses at different levels of technological sophistication. In order to achieve satisfactory quality control, the laboratory participates annually in the radon intercomparison programme of Public Health England. The study evaluates historical sets of laboratory results and assesses performance through participation in exercises. Sets of results from seven such exercises were compiled for various radon exposure levels. Measurement biases related to each exposure and type of analysis were assessed through ANOVA tests, box plots and interval plots. The statistical results indicated a general lack of performance stability over the years although they demonstrated positive signs of progress in bias decrease and a certain level of bias consistency in the various analytical approaches. The results call for further investigation of the causes of discrepancies and tendencies, aimed at continuous analysis improvements.

1. INTRODUCTION

Quality control and its continuous improvement is today a well accepted concept at many organizations who seek excellence in what they provide, whether it is a consumer item, a service or simply information [1]. Aside from the physical quality of a product, the reliability of results from an activity has become a priority in scientific fields such as the nuclear field, which has a responsibility to society when it comes to information sharing and safety of the public and the environment [2]. As one of many interests of nuclear analytical laboratories, the risk due to exposure to radon continues to drive studies on the occurrence of this naturally radioactive gas as scientific organizations (such as environmental and nuclear regulatory bodies) seek the best techniques for indoor radon monitoring at high quality levels. Thus, in today's world of facilitated knowledge exchange, collaboration among countries and laboratories in the form of intercomparison exercise programmes is a useful tool for researchers in their pursuit of measurement technique improvements and reliable results. In addition, the participation in interlaboratory exercises (with satisfactory outcomes) is a requirement for accreditation in terms of standards such as ISO/IEC 17025 [3].

The Poços de Caldas Laboratory (LAPOC) of the National Commission on Nuclear Energy has a history of indoor radon measurement studies and is currently working towards the validation of a solid state nuclear track detector (SSNTD) technique and future accreditation of its quality management system under ISO/IEC 17025. Over time, LAPOC's radon studies laboratory has developed radon monitoring projects involving passive detector analyses at different levels of technological sophistication. While one group focuses on technician supported analyses (individual and manual detector reading), the other works today with an automated nuclear track detector reading system, which provides batch analyses. In order to achieve satisfactory quality control in all of these activities, LAPOC has, since 2007, participated in the intercomparison programme of passive radon detectors organized regularly by Public Health England (PHE).

The purpose of this study was to evaluate the historical sets of LAPOC's intercomparison results and assess the laboratory's performance over the years.

2. METHODS

2.1. Public Health England intercomparison of passive radon detectors

The exercises organized by PHE consist of exposing detectors to various radon exposure levels (ranging from 50 to 3000 kBq·h m^{-3}) carried out in a 43 m^3 walk-in chamber of the static type, where radon is continuously released from dry ^{226}Ra sources. During the exercises, the radon concentration in the chamber is continuously monitored by ATMOS 12 DPX and AlphaGUARD ionization chambers, with an equilibrium factor maintained at 0.45 for all exposure levels. The uncertainty of the exposure levels is reported to be 3% at a 68% confidence level [4]. The performance evaluation conducted in this study covers exercises carried out in 2007 and 2009 (involving three levels of radon exposure) and from 2011 to 2015 (involving five levels of radon exposure). All exercises comprised a set of 10 detectors for each exposure level and a set of 10 transit detectors (whose mean value was subtracted from the measured values). Therefore, the 2007 and 2009 exercises involved 40 detector units while the other years involved 60 units. The units were pretreated and assembled in the laboratory, sent to England (where exposure took place), and were subsequently returned for etching (to reveal tracks) and counting for radon concentration determination. The measurement results were then reported to PHE for evaluation and ranking.

The current performance classification scheme applied by PHE was introduced in 2011 and is based on the determination of biased and precision errors, the results of which are applied to the calculation of the measurement error. This result is then used as a criterion for a final performance classification (rank) assigned to each set of detectors for a specific exposure level. A similar scheme was used for exercises prior to 2011, when the biased error and the standard deviation between the reported results and the reference value were calculated for each set of detectors. The sets were then ranked according to the sum of these parameters.

2.2. LAPOC's radon analysis techniques, historical data organization and performance evaluation

LAPOC has developed radon measurement techniques based on two approaches to the processes of detector etching (post-exposure treatment) and analysis:

(1) The 'manual analysis' approach involves an etching process based on detector immersion for 5.5 h in a 30% KOH etching solution at 80ºC. The detector analysis is then performed by a technician who manually counts the alpha tracks (produced by radon decay) on several images of the detector surface captured through a microscope. The number of tracks per unit area is used to calculate the radon concentration detected by the CR-39 detector material.
(2) The 'automated analysis' approach involves an etching process based on detector immersion for 1 h in a 6.25M NaOH etching solution at 98ºC. The detector analysis is then performed using an automated alpha track reading system, which detects and counts tracks and automatically converts this information to radon concentration.

For both approaches, the pre-exposure treatment (immersion in liquid detergent solution, drying) and dosimeter assembly is similar.

Sets of results of both manual and automated analyses conducted for the intercomparisons during the years 2007–2015 were arranged according to exposure level ranges defined by PHE as reference values (see Table 1). In line with the objective of this work (performance evaluation) and because of the differences between the two analysis approaches, the exercise participants were separated into manual (Lab 1) and automated (Lab 2) analysis groups. Lab 1 participated in all exercises (2007–2015), while Lab 2 participated only in the 2013, 2014 and 2015 exercises.

TABLE 1. REFERENCE EXPOSURES

Exposure level code	Exposure (kBq·h m^{-3})						
	2007	2009	2011	2012	2013	2014	2015
A	140	165	112	138	144	132	145
B	255	330	382	438	347	327	330
C	–	–	902	717	657	630	719
D	–	1436	1516	1487	1326	1477	1353
E	1913	–	2174	2385	2990	2382	2259

Although PHE's intercomparison performance assessment is achieved using the mean of each set of 10 detectors for each of the three or five exposure levels, the evaluation proposed here considered the results from each detector individually. Following the organization of results by year of participation, levels of exposure and type of analysis, the percentage biased error was calculated for each detector individually. Spreadsheets categorized by exposure levels were then created to enable ANOVA tests to be applied to the bias error data using the software MiniTab. With this arrangement, the bias means related to each year at each level of radon exposure could be compared with each other. In this evaluation by exposure level, the year becomes the variable since the aim of the study was to evaluate progress over time. In addition, box plot graphs and interval plots were produced, related to exposure level. Finally, a 95% confidence interval was determined for all tests.

3. RESULTS AND DISCUSSION

For both manual and automated analysis approaches, the results obtained from the ANOVA tests demonstrated a lack of bias consistency over the years, with all the resulting means observed being statistically different across the various exposure levels. Despite this lack of consistency, Lab 1 (manual analysis) displayed a level of consistency in its bias throughout the seven year exercise period (see Fig. 1), while Lab 2 (automated analysis) demonstrated significant progress in reducing bias (dispersion and percentage) throughout its three year exercise period (see Fig 2). Significant differences between the two analysis approaches were observed when manual and automated analysis result biases were compared, as illustrated in Fig. 3.

Upon specific evaluation of Lab 2's performance in the 2015 exercise, which resulted in negative bias values for all five exposure levels (mean values) in addition to indicating a growth in bias value (in module) as radon exposure levels increased from A to E, a correction of the measured values through linear regression was considered. The concept was applied to the set of five means obtained from the 2015 exercise. The results are shown in Fig. 4.

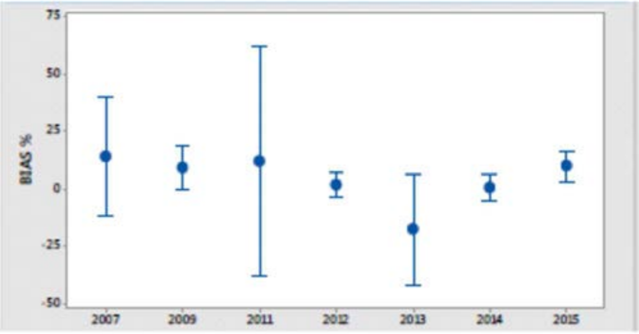

FIG. 1. Bias interval plot, exposure level A (Lab 1, manual analysis).

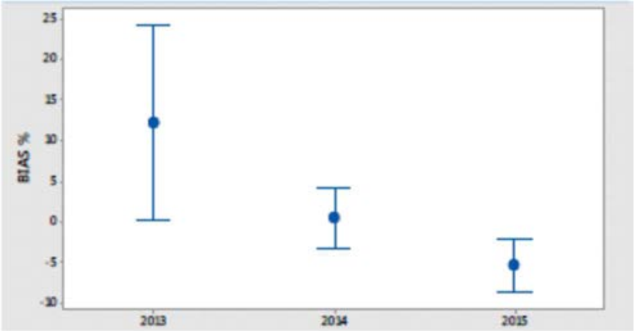

FIG. 2. Bias interval plot, exposure level B (Lab 2, automated analysis).

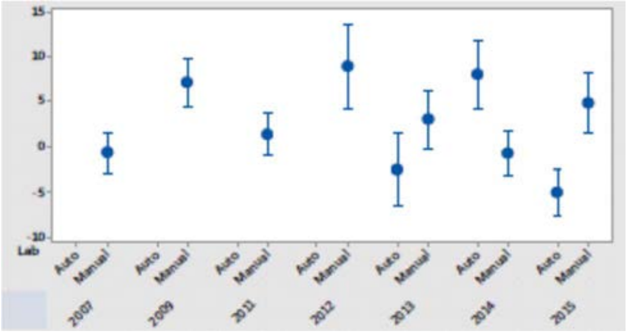

FIG. 3. Bias interval plot, exposure level C, comparing Labs 1 and 2 in 2013, 2014 and 2015.

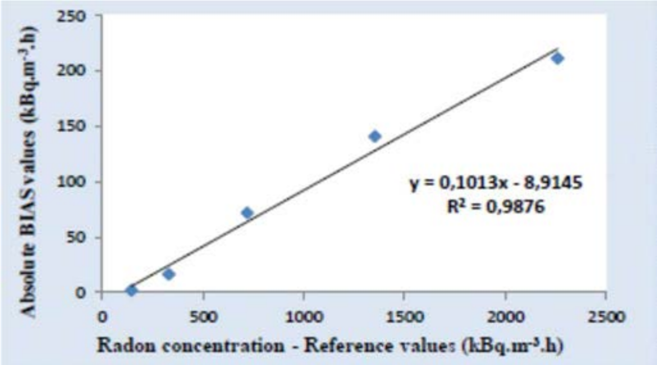

FIG. 4. Measurement correction curve, 2015 intercomparison exercise (automated analysis).

4. CONCLUSIONS

The statistical data generated by the ANOVA tests applied to the intercomparison result bias values generally indicate a lack of performance stability over time when judged in terms of quality control practices. The two different analysis approaches led to significantly different results. Moreover, consistency was not observed within each exposure level. However, progress in bias decrease over a three year period (Lab 2, automated analysis) and a demonstration of bias consistency over seven exercises (Lab 1, manual analysis) are regarded as positive outcomes of the historical progress of performance. Further investigations are needed on the feasibility of applying a correction to the biased measurement results that could lead to the improvement of the automated analysis method. The results also call for further investigation into the causes of the observed discrepancies and tendencies, with a view to identifying corrections leading to analysis improvements, in line with the philosophy of quality control systems.

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Poster Presentation

GEORAD: GEO-REFERENCED DATABASE OF ENVIRONMENTAL RADIOACTIVITY IN BRAZIL

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Abstract

GEORAD is a database of geo-referenced samples containing information dealing with radioactivity in the environmental media of Brazil. The main objectives of the GEORAD project are to map the distribution of environmental radioactivity in Brazil, to compile and assemble the data in a database, to attach the data to a geographic information system and to provide access to researchers of sample data via the website. The system provides information on the concentrations of terrestrial radionuclides of natural origin, cosmogenic radionuclides and fallout radionuclides in samples of soil, water and food, among others, along with the geographical locations of the samples, which are identified on a map of Brazil. A spreadsheet containing all the data and information about the sample can be also obtained. The input data were obtained from the literature or authors in the field, or was provided directly by researchers. As a result, the database system can enable the available data to be utilized for further research. The system also provides reference information on the source of the data information, enabling the data to be referenced. The system can provide important aspects of the spatial information via the software GoogleEarth™, as well as the radionuclide concentrations and the digital object identifier of the scientific article used as reference for the sample data collected. This system is available to the public via the homepage of the Institute of Radiation Protection and Dosimetry, requiring only a simple registration.

1. INTRODUCTION

The main objectives of the GEORAD project are to map the distribution of environmental radioactivity in Brazil, to compile and assemble the data in a database, to attach the data to a geographic information system and to provide access to researchers of sample data via the website. The system provides information on the concentrations of terrestrial radionuclides of natural origin, cosmogenic radionuclides and fallout radionuclides in samples of soil, water and food, among others, along with the geographical locations of the samples, which are identified on a map of Brazil. A spreadsheet containing all the data and information about the sample can be also obtained. As a result, the database system can enable the available data to be exploited to its maximum potential for further research and to allow new research on existing information. The system also provides reference information on the source of the data information, enabling data citation and linkage with publications to increase visibility and accessibility of the data and the research itself.

2. METHOD

The methodology is based on the ranking of samples according to type and/or use, therefore enabling the systematic storage and standardization of research in the database. The methodology applied in the project allowed the prioritization of data to simplify its cataloguing, searching for information and localizing geo-referencing of the samples. This led to normalization of tables in the database [1]. Each sample was associated with a considerable amount of information that guided the search among the various types of samples contained in the database. These samples were classified according to the following criteria for environmental monitoring [2]:

(a) Radionuclides: natural, cosmogenic and fallout;
(b) Geographic location: region, state, county and local levels;
(c) Environmental compartment: sample class and type or use the sample;
(d) Source of data: bibliographic reference, supplied directly by the institution concerned.

3. RESULTS

The GEORAD system is available at http://georad.ird.gov.br in Portuguese, Spanish and English. When the database is accessed by the user, it is possible to perform a systematic search based on the methodology developed in this project, using catalogued samples. Information about the Brazilian regions, states, cities and classes and types of radionuclides can be chosen for more detailed research. After searching the samples, the system shows a list containing the selected data. The user can see immediately the number of samples available, as well as their location and their classification. Important aspects of spatial information can be obtained via the software GoogleEarth^{TM}, where the user can gain access to information on a given sample, namely, its geographical location shown on a map of Brazil, information on the concentrations of radionuclides and data on the digital object identifier of the scientific article used as the reference for the sample data collected. The GEORAD system is continuously expanded and updated and currently contains data from more than 2000 samples.

4. CONCLUSIONS

The GEORAD system is based on a survey of published data in the scientific literature and data provided directly by researchers. It has enabled dissemination to the public of data on radioactivity in the environment and has promoted the emergence of a new family of products based on the same technology (G-Mapping of Soils and G-Radiation Facilities). The GEORAD system achieves the following goals:

(a) To allow systematic and secure storage of data generated on radioactivity;
(b) To generate facilities for decision making in the field of new research;
(c) To provide information on radioactivity data in the regions of Brazil;
(d) To allow studies of correlations and trends;
(e) To support the interpretation of data, allowing comparisons.

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Poster Presentation

SHORT TERM TEMPERATURE STABILITY TEST FOR URANIUM SOIL REFERENCE MATERIAL

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Abstract

A soil material produced by the Metrology Department of the Institute of Radiation Protection and Dosimetry is a candidate reference material for chemical analysis of natural uranium with metrological traceability to be used in environmental analysis, equipment calibration, method validation and quality control. This paper describes one important step to produce a soil reference material for natural uranium. In the production of soil reference materials, the short term stability is an essential property to be verified in order to ensure that the basic characteristics of the material will remain constant for a specific period. A methodology to determine the soil short term stability after the production steps in laboratory is described. The test performed in this study used neutron activation analysis for uranium determination in soil samples. The chosen temperatures to determine the short term stability were 20°C and 60°C, and the slopes and their uncertainties were obtained from the regression lines. The control samples were maintained at a temperature of –20°C and the t-test was applied. At 20°C the t-value obtained was 0.34 and the critical value was 2.78. At 60°C the t-value was 1.19 and the critical value was 3.18. Since in both cases the calculated t-value is lower than the critical value, it can be concluded with 95% confidence that no significant changes in uranium concentration in the soil occurred during the period studied at temperatures of 20°C and 60°C, showing stability at these temperatures.

1. INTRODUCTION

Brazil has large deposits of uranium. Radioanalytical procedures to determine uranium concentrations in natural matrices are critical for an accurate assessment of the concentration of this element in the various minerals of interest. The environmental control of the radionuclide concentration in the vicinity of uranium mines is another area which also requires reliable measurements and standards. For this reason, the Metrology Laboratory (LNMRI) of the Institute of Radiation Protection and Dosimetry (IRD) developed studies to prepare matrix reference materials and conduct laboratory intercomparison exercises. In recent years the IRD has also begun to regularly conduct a proficiency test ‘Programa Nacional de Intercomparação’ (PNI). The PNI is conducted to support laboratories in Brazil involved in radionuclide analysis of environmental samples. They are involved in various fields of application and cover a wide range of objectives of regional and interregional projects. To cover the vast range of requirements and the increasing demand for PNI, the Reference Materials Group at the IRD laboratory uses either natural matrix reference materials or matrix materials spiked with calibrated standard solutions as reference material. The characterization of natural matrix reference materials is expensive and time consuming. Despite all efforts, the availability of matrix reference materials remains limited and it is not always possible to meet the target criteria for a specific proficiency test, e.g. type of matrix, concentration of analytes, total combined uncertainty associated with the target values. Radioanalytical determinations require standards not only for instrument calibration but also for the evaluation of the analysis performance. Therefore the production of a uranium soil reference material is extremely important to all programmes that involve uranium determination.

2. METHOD

2.1. Sample collection and preparation

About 600 kg of wet material from the ground on a farm in Pocos de Caldas, Minas Gerais, (21°55'47.9"S, 46°35'13.2"N) were collected to a depth of around 50 cm. The collected soil was taken to IRD/LNMRI and was taken through several steps. The raw soil material was initially cleaned by hand with the removal of stones, roots, leaves and other macroscopic impurities. The material was then dried in a laboratory oven with air flow at 500°C overnight. The material was then sieved to separate all the components larger than 3 mm. The presence of small pieces of vegetable matter mixed with the soil necessitated a procedure for the elimination of organic matter by calcination in a furnace at 4500°C for 24 h. The soil, now free of organic matter, was milled in a Retsch PM400 ball mill (see Fig. 1), passed through a 75 µm sieve (see Fig. 2), homogenized (see Fig. 3), submitted to an automatic bottle filling facility and sterilized with ^{60}Co gamma radiation. After these steps, 120 kg of processed material (see Fig. 4) were obtained from the original 600 kg collected.

FIG. 1. Ball mill.

FIG. 2. Sieve system.

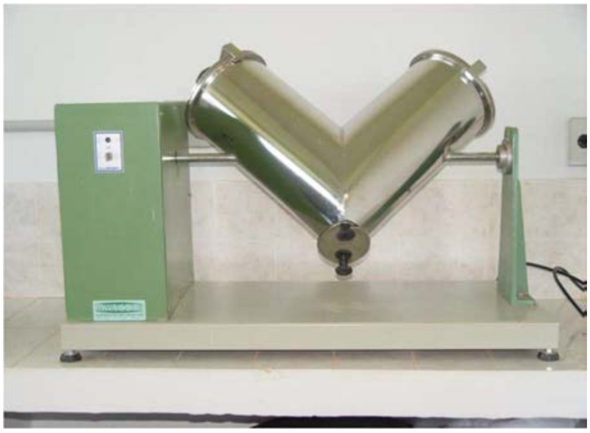

FIG. 3. Type V homogenizer.

FIG. 4. The final soil package.

2.2. Determination of the stability of the uranium concentration

Samples for the stability tests, including three control samples, were randomly selected from the 120 kg of processed soil and placed in vials. The samples for determining the short term stability were maintained at 20°C in a controlled weighing room with an air conditioning system or at 60°C in a laboratory oven. The control samples were kept at a temperature of –20°C. The uranium concentrations in the samples were measured by neutron activation analysis. This was conducted in the research reactor IEA-R1 of Instituto de Pesquisas Energéticas e Nucleares, São Paulo using an irradiation time of 8 h and a thermal neutron flux in the range 6.5×10^{12} to 4.5×10^{12} $n\cdot cm^{-2}\cdot s^{-1}$. The induced gamma activity was measured using a POP TOP model high purity germanium detector (EG&G ORTEC) with a resolution of 1.90 keV for the 1332.49 keV ^{60}Co peak, with associated electronics connected. The samples for the stability tests were measured repeatedly at time intervals of 7, 14, 21, 28 and 60 d. The measured uranium concentrations were divided by the average uranium concentration of the control samples obtained at –20°C to give Qt ratios. These ratios were plotted against time and the regression lines obtained. The t-test was then applied using the slope and uncertainties obtained from the regression lines.

3. RESULTS

The average uranium concentration measured in the control samples was 69.7 mg/kg with a standard deviation of 2.4 mg/kg. The uranium concentrations measured in the stability tests are shown in Table 1. The Qt ratios are shown plotted against time in Figs 5 and 6.The regression lines can be represented by Eqs (1) and (2).

$$y = -0.0004\,x + 0.9982 \qquad (20°\mathrm{C}) \tag{1}$$

$$y = 0.0008\,x + 0.9428 \qquad (60°\mathrm{C}) \tag{2}$$

The t-value at 20°C was 0.34, while the critical value was 2.78. The t-value at 60°C was 1.19, while the critical value was 3.18.

TABLE 1. URANIUM CONCENTRATION (mg/kg) AT 20°C AND 60°C

Elapsed time (d)	Uranium concentration (mg/kg)	
	20°C	60°C
7	71.3 ± 0.3	65.5 ± 0.2
14	71.0 ± 0.4	68.7 ± 0.4
21	65.5 ± 0.4	66.5 ± 0.4
28	66.0 ± 0.3	–
60	69.6 ± 0.3	68.9 ± 0.3

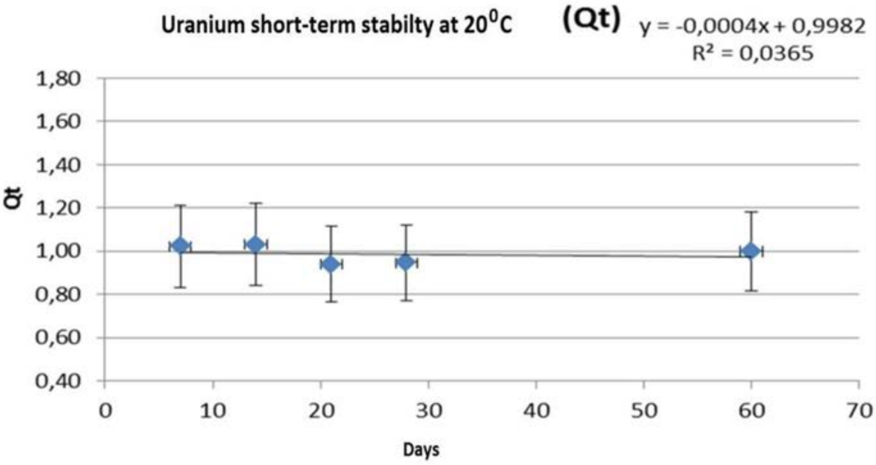

FIG. 5. Variation of the ratio Qt with time at 20°C.

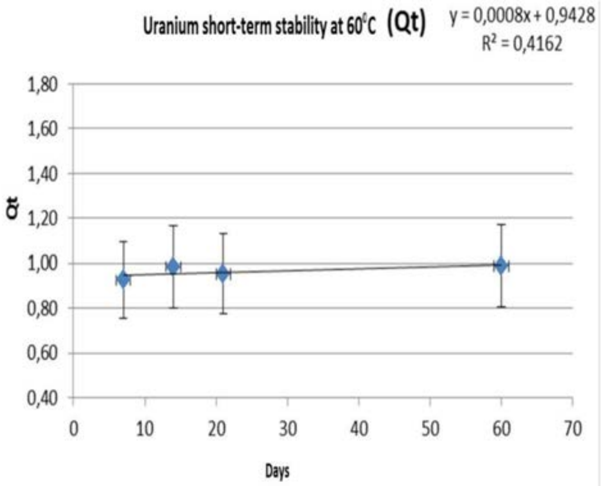

FIG. 6. Variation of the ratio Qt with time at 60°C.

4. CONCLUSION

Since in both cases the calculated t-value is less than the critical value, it can be concluded with 95% confidence that no significant changes in uranium concentration occurred at 20°C or 60°C, demonstrating that the uranium concentrations in the soil samples prepared as reference material were stable in the short term at these temperatures.

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Poster Presentation

UNCERTAINTY ESTIMATION USING THE SPREADSHEET APPROACH FOR DETERMINATION OF ^{210}Po IN ENVIRONMENTAL SAMPLES BY ALPHA SPECTROMETRY

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Abstract

Uncertainty measurements provide information about the confidence of results as well as the dispersion of the values attributed to the results. The correct calculation of uncertainties offers effective quality assessment in and between laboratories and helps stakeholders in decision making processes. Since ^{210}Po is used for a variety of purposes, such as radiological impact assessment, as a tracer of environmental processes and as an indirect determinant of its progenitor ^{210}Pb, it is important to correctly express the uncertainties in its activity concentration. This paper demonstrates the use of the spreadsheet method to estimate uncertainties of ^{210}Po activity concentration in IAEA Certified Reference Material IAEA–477.

1. INTRODUCTION

Among naturally occurring radioactive elements in the environment, polonium plays an important role since it is one of the most toxic, giving rise to risks to humans through direct uptake, ingestion, or inhalation [1, 2]. Polonium-210 belongs to the ^{238}U decay series and its fate depends on other members of this series including ^{226}Ra, ^{222}Rn and ^{210}Pb [3]. The determination of Po activity in sediment samples through alpha spectrometry analysis involves drying the sediment and radiochemical separation of the ^{210}Po. All potential losses of the analyte during the process can be verified through the addition of a ^{209}Po tracer of known activity.

2. RADIOCHEMICAL METHOD

The IAEA sample was prepared using about 1 g of the dried material and 10 mL of ^{209}Po tracer. Both were transferred to a Teflon beaker that was kept under agitation for 20 min for correct homogenization. The sample was then digested in an open system with mineral acids. The residue was dissolved using 1.5M HCl and filtered. The filtered solution was transferred to a deposition cell using 1.5M HCl. In order to eliminate interferences, 1 g of ascorbic acid was added. The ^{210}Po was deposited onto a silver planchet under agitation for 4 h at 80°C. Finally, the planchet was counted on a surface barrier detector in an alpha spectrometry system.

3. THE SPREADSHEET APPROACH

The spreadsheet approach [4], which is based on the uncertainty propagation law, requires the use of equations to calculate the result, the parameter values and their associated uncertainties. A detailed description of the method and an example are given in Ref. [5]. The ^{210}Po activity is calculated using Eq. (1), the ^{210}Po activity concentration is calculated by Eq. (2) and the correction factors f_1, f_2, f_3 and f_4 are given by Eqs (3–6).

$$A_{\mathrm{A}} = C_{\mathrm{T}} V_{\mathrm{T}} \left(\frac{R_{\mathrm{GA}} - R_{\mathrm{BA}}}{R_{\mathrm{GT}} - R_{\mathrm{BT}}} - q_1 \right) \cdot \left(\frac{P_{\alpha\mathrm{T}}}{P_{\alpha\mathrm{A}}} \right) \tag{1}$$

$$\alpha_{\mathrm{A}} = \frac{A_{\mathrm{A}}}{m_{\alpha} q} f_1 f_2 f_3 f_4 \tag{2}$$

$$f_1 = \exp\left(+ \lambda_{\mathrm{A}} \left(t_{\mathrm{s}} - t_{\mathrm{e}} \right) \right) \tag{3}$$

$$f_2 = \frac{\lambda_{\mathrm{A}} t_{\mathrm{g}}}{1 - \exp\left(- \lambda_{\mathrm{A}} t_{\mathrm{g}} \right)} \tag{4}$$

$$f_3 = \exp\left(- \lambda_{\mathrm{T}} \left(t_{\mathrm{s}} - t_{\mathrm{c}} \right) \right) \tag{5}$$

$$f_4 = \frac{\lambda_{\mathrm{T}} t_{\mathrm{g}}}{1 - \exp\left(- \lambda_{\mathrm{T}} t_{\mathrm{g}} \right)} \tag{6}$$

where A_{A} is the activity of ^{210}Po,
α_{A} is the activity concentration of ^{210}Po,
m_{α} is the sample mass,
q is the wet and ash rate,
f_1 is the decay correction from the end of sampling to the beginning of measurement,
f_2 is the decay correction during counting,
f_3 is the decay correction for the tracer from calibration to the measurement,
f_4 is the decay correction for the tracer during measurement,
C_{T} is the tracer concentration at calibration,
V_{T} is the tracer volume,
R_{GA} is the gross counting rate of ^{210}Po,
R_{BA} is the blank counting rate of ^{210}Po,
R_{GT} is the gross counting rate of the tracer ^{209}Po,
R_{BT} is the blank counting rate of the tracer ^{209}Po,
q_1 is the tracer impurity,
$P_{\alpha\mathrm{T}}$ is the sum of emission probabilities of the tracer lines contributing to count rate,
$P_{\alpha\mathrm{A}}$ is the sum of emission probabilities of the analyte lines contributing to count rate
λ_{A} is the ^{210}Po decay constant,
λ_{T} is the ^{209}Po decay constant,
t_{s} is the start of counting,
t_{e} is the end of sampling,
t_g is the gross counting time,
t_{c} is the tracer calibration time.

The uncertainties of R_{GA}, R_{BA}, R_{GT}, R_{BT}, f_1, f_2, f_3 and f_4 are calculated according to the equations presented in Ref. [6], while the uncertainties of q_1, $P_{\alpha T}$ and $P_{\alpha A}$ were considered negligible.

The radiochemical yield is not directly used in the activity concentration calculation but it gives an indication of the performance of the chemical and source preparation procedure. Equation 7 shows the calculation, where ε represents the efficiency of the detector for alpha particles.

$$\eta = \frac{R_{GT} - R_{BT}}{\varepsilon \, C_T V_T} \tag{7}$$

4. RESULTS

Table 1 shows the parameters, their values and respective uncertainties for sample 1, from IAEA-447 Certified Reference Material. The calculated ^{210}Po activity concentration using Eq. (1) was found to be 437 Bq/kg.

TABLE 1. SUMMARY OF PARAMETERS, VALUES AND UNCERTAINTIES FOR SAMPLE 1

Parameter	Value	Uncertainty
m_a	0.0009951 kg	± 0.0000001 kg
v_T	9.9963 mL	±0.0001 mL
R_{GA}	0.06061470	±0.00055046
R_{BA}	6.9986×10^{-5}	$\pm 1.8704 \times 10^{-5}$
R_{GT}	0.0351	±0.0004
R_{BT}	0.0072035	±0.0001898
q_1	0	0
$P_{\alpha T}$	1	0
$P_{\alpha A}$	1	0
C_T	0.02204 Bq/mL	± 0.00012 Bq/mL
q	1	0
λ_A	5.7976234×10^{-8} s^{-1}	± 7.1225780×10^{-9} s^{-1}
λ_T	1.91×10^{-10} s^{-1}	± 2.16×10^{-11} s^{-1}
t_s–t_e	172800 s	± 1 s
t_g	200042.4 s	± 1 s
t_s–t_c	5 710 176 000 s	± 1 s
f_1	1.01007	± 0.00124
f_2	1.005810	± 0.000715
f_3	0.897	± 0.013
f_4	1.000019	± 0.000002

In the spreadsheet, the parameters were listed vertically and their uncertainties horizontally. The columns were completed with the values of each parameter. First, in each line, the uncertainty was added to the value of the parameter, adjusting one value in each column (see Table 2). For example in the second line, the value of 9.9963 was adjusted to 9.9964 in the second column, which represents the volume of tracer plus its uncertainty. Secondly, the activity concentration was recalculated for each column, giving the adjusted result. Next, the difference between the changed result and the 'real' result was calculated for each parameter, giving the residual values (see Table 2). Then, the residuals were squared for each column and were summed up for all the parameters. The sum value was 101.83. Finally, the uncertainty was found by taking the square root of the sum of squares, obtaining a final activity concentration value of 437 ± 10 Bq/kg.

The parameters that contributed most to the final uncertainty, together with their percentage contributions, were as follows:

R_{GA}	15.53%,
R_{BA}	0.02%,
R_{GT}	41.09%,
R_{BT}	8.80%,
C_T	5.57%,
f_1	0.28%,
f_2	0.10%,
f_3	28.60%.

5. DISCUSSION

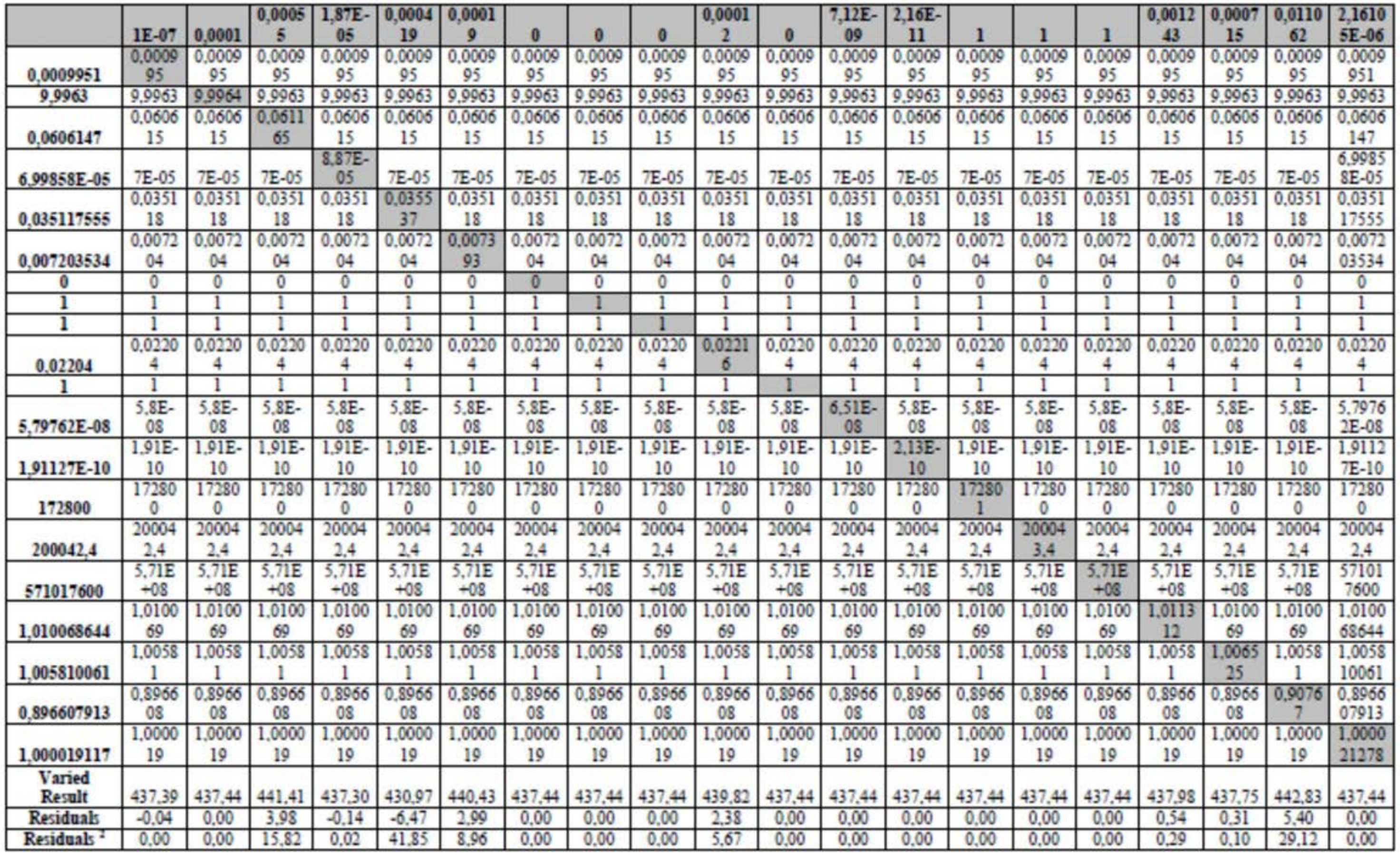
Considering the activity value for sample 1 specified on the reference sheet, it was expected to find an activity concentration of 423 ± 10 Bq/kg. The result shows an activity concentration of 437 ± 10 Bq/kg, confirming that the polonium analysis and the uncertainty measurement are reliable. Considering a detector efficiency of 0.1855 for alpha particles, the radiochemical yield was 68.3%.

According to the percentage contributions, the main contributors to the final uncertainty are the gross counting rate of the tracer R_{GT} (41.1%) and the decay correction factor of the tracer f_3 (28.6%). The gross counting rate depends on the peak area of the tracer and the gross counting time. Therefore, an increase in the counting time can result in an improvement in the results. On the other hand, the decay correction factor depends on the ^{209}Po decay constant and the time between the tracer calibration and the beginning of measurement. This contribution could be minimized by using a newer tracer, since it would decrease the parameter $t_s – t_c$ and consequently its uncertainty.

6. CONCLUSION

The spreadsheet approach has shown a satisfactory performance in the calculation of uncertainties for ^{210}Po activity concentration, resulting in reliable and accurate values. Furthermore, it has helped to identify which parameters contribute more to the overall uncertainty. As a consequence, it was possible to recognize where more effort could be made in order to decrease uncertainties in future analyses. Finally, this step by step demonstration of the spreadsheet approach can offer suitable information for the calculation of activity concentration of environmental samples by alpha spectrometry.

TABLE 2. CALCULATION OF ADJUSTED RESULTS, RESIDUALS AND SQUARED RESIDUALS



	1E-07	**0,0001**	**0,00055**	**1,87E-05**	**0,000419**	**0,00019**	**0**	**0**	**0**	**0,00012**	**0**	**7,12E-09**	**2,16E-11**	**1**	**1**	**1**	**0,001243**	**0,000715**	**0,011062**	**2,16105E-06**
0,0009951	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,000995	0,0009951
9,9963	9,9963	9,9964	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963	9,9963
0,0606147	0,060615	0,060615	0,061165	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,060615	0,0606147
6,99858E-05	7E-05	7E-05	7E-05	8,87E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	7E-05	6,99858E-05
0,035117555	0,035118	0,035118	0,035118	0,035118	0,035537	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035118	0,035117555
0,007203534	0,007204	0,007204	0,007204	0,007204	0,007204	0,007393	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007204	0,007203534
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02216	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204	0,02204
1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
5,79762E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	6,51E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,8E-08	5,79762E-08
1,91127E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	2,13E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91E-10	1,91127E-10
172800	172800	172800	172800	172800	172800	172800	172800	172800	172800	172800	172800	172800	172800	172801	172800	172800	172800	172800	172800	172800
200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200042,4	200043,4	200042,4	200042,4	200042,4	200042,4	200042,4
571017600	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	5,71E+08	571017600
1,010068644	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,010069	1,011312	1,010069	1,010069	1,010068644
1,005810061	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,00581	1,006525	1,00581	1,005810061
0,896607913	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,896608	0,90767	0,896607913
1,000019117	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000019	1,000021278
Varied Result	437,39	437,44	441,41	437,30	430,97	440,43	437,44	437,44	437,44	439,82	437,44	437,44	437,44	437,44	437,44	437,44	437,98	437,75	442,83	437,44
Residuals	-0,04	0,00	3,98	-0,14	-6,47	2,99	0,00	0,00	0,00	2,38	0,00	0,00	0,00	0,00	0,00	0,00	0,54	0,31	5,40	0,00
Residuals ^{2}	0,00	0,00	15,82	0,02	41,85	8,96	0,00	0,00	0,00	5,67	0,00	0,00	0,00	0,00	0,00	0,00	0,29	0,10	29,12	0,00

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Poster Presentation

CONCENTRATIONS OF ^{226}Ra, ^{232}Th AND ^{40}K IN BRAZILIAN WALL PAINT

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Abstract

Geological materials used as building materials are a source of radiation exposure due to the presence of radionuclides of natural origin. Wall paint is one of the building materials to be considered for radiological evaluation as it generally contains titanium dioxide pigment obtained from minerals such as ilmenite and rutile which contain ^{238}U and ^{232}Th series radionuclides and ^{40}K. In this work, radionuclide concentrations were determined in 50 commercial Brazilian white latex wall paints using high resolution gamma-ray spectrometry. The following activity concentrations were measured: 1.41–38.7 Bq/kg (^{226}Ra), 0.9–101.2 Bq/kg (^{232}Th) and 5.9–256 Bq/kg (^{40}K). These results demonstrate that the wall paints studied in this work are safe for use.

1. INTRODUCTION

Radionuclides of natural origin with half-lives of the same order as that of the age of the Earth (i.e. radionuclides in the ^{238}U and ^{232}Th series and ^{40}K) are a ubiquitous source of human exposure [1, 2]. Workplaces and homes have implications for exposure because significant periods of time are spent inside them — it is not uncommon for some individuals to spend 80% of their time in homes and virtually every building material contains measurable concentrations of ^{226}Ra, ^{232}Th and ^{40}K. Knowledge of the radionuclide concentrations in building materials is needed for any assessment of population exposure. Several studies have been conducted worldwide to evaluate radioactivity in building materials such as rock, granite, marble and sand [3–7] but it would appear that no studies have been performed to determine activity concentrations in internal or external wall paints.

The basic raw materials for the production of almost all types of paint are resin, pigment, solvent and additives. The pigment gives the paint its colour, solvents make it easier to apply the resin and promote drying, while additives are used as fillers, antifungal agents, etc. The most common pigment material, titanium dioxide, provides whiteness and opacity. Titanium dioxide is a simple inorganic compound obtained from ilmenite and (to a lesser extent) rutile in heavy-mineral sand and accounts for 92% of the worldwide demand for titanium minerals [8]. Since ilmenite sand contains up to 500 ppm thorium and lesser amounts of uranium [9], it is easy to suspect that wall paints, like other construction materials, could contain significant levels of radioactivity [10]. Several studies have been conducted to evaluate the occupational exposure of titanium production workers [10–13] but public exposure from wall paints used in buildings seems not to have been investigated. In Brazil, the relative uses of titanium dioxide are 85.5% for paint, 8.6% for the steel industry, 6.4% for iron alloys, 1.6% for electrodes and 0.8% for floors and tiles [14]. Brazil is one of the world's top five markets for coatings, and manufactures paints for a variety of applications. There are hundreds of large, medium and small manufacturers spread throughout the country. The top ten manufacturers account for 75% of total sales [15].

2. EXPERIMENTAL

Samples of 50 brands of wall paint from various manufacturers were each sealed tightly in a 100 mL HDPE flat bottomed cylindrical flask with a screw cap and bubble spigot and stored for approximately 4 weeks in order to ensure radioactive secular equilibrium [16]. All samples were measured for 150 000 s with a coaxial extended range high purity germanium detector of 25% relative efficiency, with conventional electronics and an EG&G ORTEC Spectrum Master 919 4-k multi-channel analyser. The spectra were analysed using WinnerGamma software [17]. Background radiation was determined by measuring an ultra-pure water sample and the detector efficiency curve was determined with a multi-element standard aqueous radioactive solution sample, both in the same geometry of the samples. The activity concentration of ^{40}K was calculated from its single gamma energy peak of 1461 keV. The activity concentration of ^{226}Ra was determined by the weighted mean of the concentrations derived from the ^{214}Pb and ^{214}Bi gamma peaks and the activity concentration of ^{232}Th was determined by the weighted mean of the concentrations derived from the ^{212}Pb, ^{212}Bi and ^{228}Ac gamma peaks [16]. All activity concentrations were determined with self-attenuation corrections [18]. The apparent densities of the samples varied from 0.97 ± 0.03 to 1.46 ± 0.04 g/cm^3.

3. RESULTS AND DISCUSSION

The activity concentrations for all the radionuclides considered are shown in Fig. 1 for each paint sample. The activity concentrations of ^{232}Th were generally higher than those of ^{226}Ra. The correlation between ^{232}Th and ^{226}Ra in the samples is shown in Fig. 2 for each paint sample. Sample E08 (the outlier point in Fig. 2) shows a different behaviour from all the other samples, suggesting that its ^{226}Ra activity concentration is influenced by a constituent other than titanium dioxide.

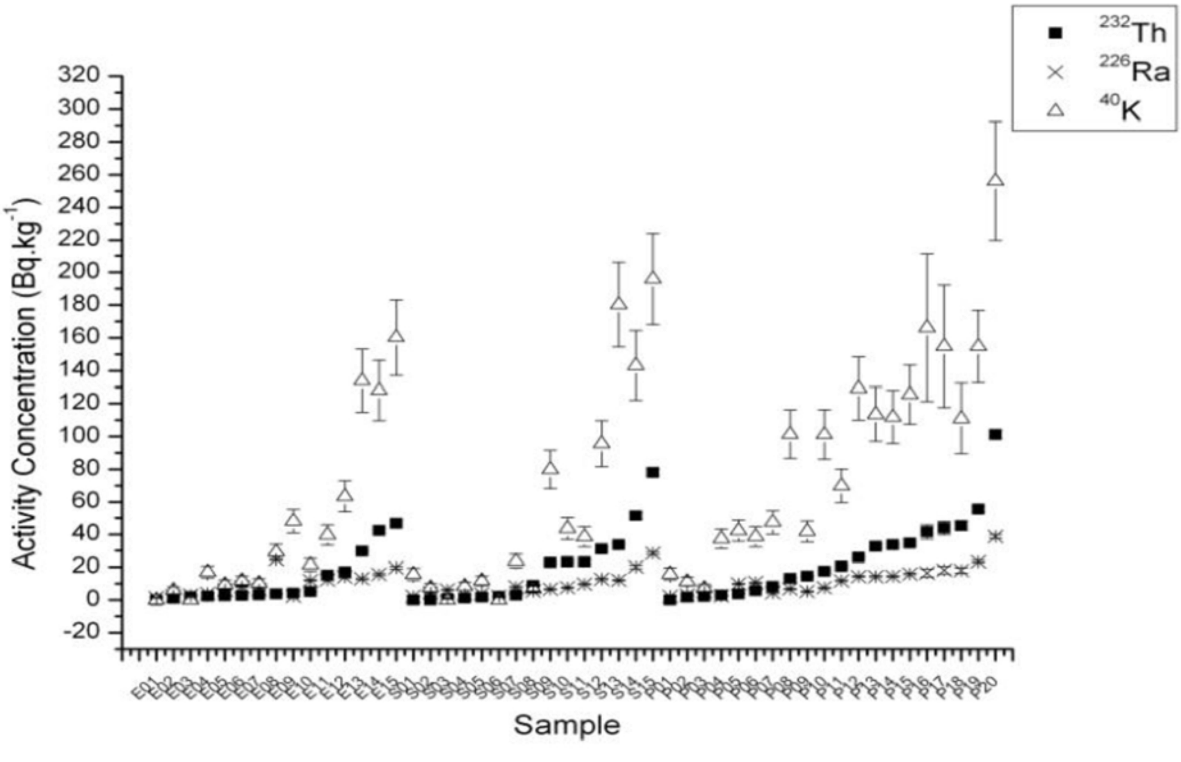

FIG. 1. Radionuclide activity concentrations in samples of 50 different wall paints. The paints are labeled E, S and P for economic, standard and premium quality, respectively.

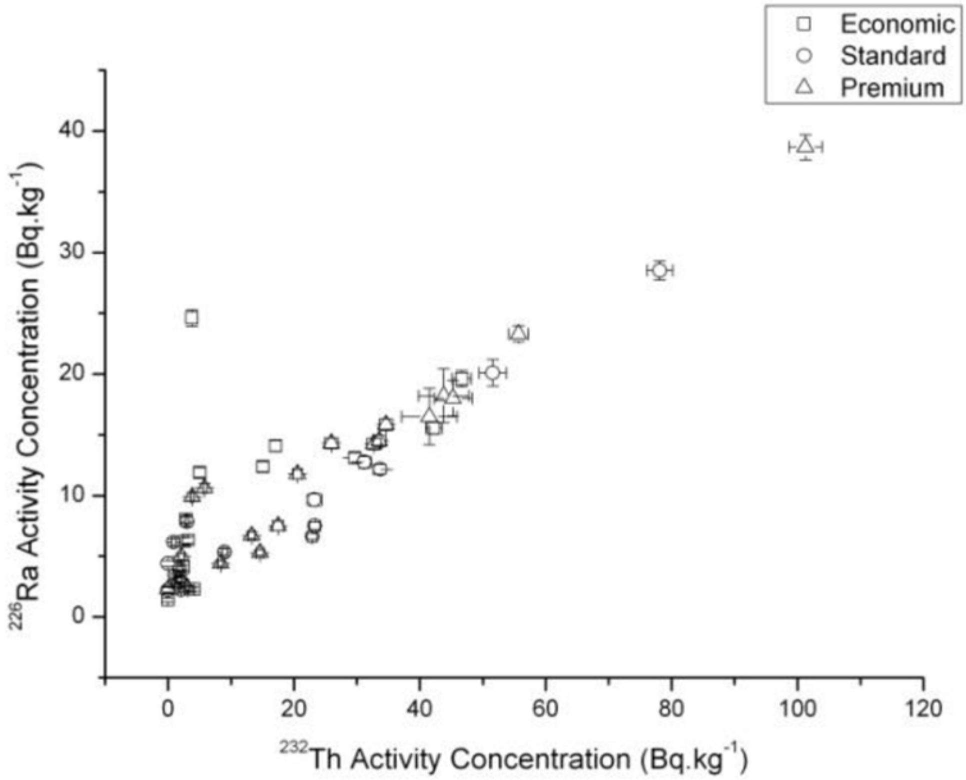

FIG. 2. Correlation between ^{232}Th and ^{226}Ra activity concentrations.

4. CONCLUSIONS

The results of the activity concentration measurements in 50 samples of wall paint show elevated ^{232}Th concentrations in most samples, as would be expected for a material originating from ilmenite or rutile. The maximum activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were 38.7 ± 1.0, 101.2 ± 2.6 and 260 ± 40 Bq/kg, respectively, all in sample P20. No correlation between the activity concentration and the grade of wall paint (economic, standard or premium) was observed. It was concluded that all paints were safe for use.

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Poster Presentation

SPATIAL SURVEY OF RADIOACTIVITY IN BRAZIL

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** Geological Survey of Brazil

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Abstract

A survey of natural radioactivity and ^{137}Cs is being performed throughout Brazil. Thousands of samples are being collected in all Brazilian States. The main objective is to increase the knowledge of the distribution of natural radioactivity in the soil of Brazil and to provide (among other things) essential information for epidemiological and geological studies, for environmental radiation safety research and for decision making processes regarding soil contamination levels. Soil samples are being collected by the Geological Survey of Brazil and sent to the Institute of Radiation Protection and Dosimetry, where they are processed and analysed for determination of radionuclide concentrations by gamma spectrometry. A database has been designed containing all the sample information to be made available to the scientific community by internet tools. This paper describes the project design, the methodology adopted for sample handling and storage, the analytical quality control, a national map containing the location of the collected samples and the main results obtained to date.

1. INTRODUCTION

Human exposure to radioactivity is unavoidable, owing to the ubiquity of radionuclides of natural origin in the environment. Radionuclides of natural or anthropogenic origin occur in all environmental compartments, including rocks and soil. Radionuclides are present at various concentrations in soil worldwide according to the types of rock from which the soil originated [1]. Studies of the distribution of radioactivity in soil have been carried out in many countries with the objective of evaluating public exposure, developing radiation maps and verifying changes in environmental radioactivity caused by nuclear, industrial and other human activities.

In Brazil, measurements have been made of the concentrations of ^{238}U, ^{226}Ra, ^{228}Ra (^{232}Th), ^{40}K and ^{137}Cs in soil samples collected from some Brazilian regions [2–7]. Most of these studies were restricted to certain locations, performed in regions of high background, or designed to meet specific research goals (e.g. soil to plant uptake, soil erosion). A systematic survey of environmental radioactivity had not previously been performed in Brazil. Therefore, the MAPRAD project was initiated, aimed at systematically measuring radioactivity in soil and its contribution to the annual effective dose received by members of the public, and providing information for epidemiological and geological studies. A further objective was to make the data available to the public in the form of an online and geo-referenced database of the activity concentrations of radionuclides in surface horizons. However, it is not adequate to provide only a database — besides the challenge of performing spatial mapping, there are the challenges of ensuring the traceability of the stored samples and the quality of data produced by the different laboratories. These are all requirements that the MAPRAD project was designed to meet. This paper presents the MAPRAD project and the methodology adopted in it.

2. MATERIALS AND METHODS

The Geological Survey of Brazil (CPRM) has been developing the Multi-purpose Geochemical Survey Project (PGAGEM) since 2008, in order to assess the chemical composition of the bedrock, soil, surface water and groundwater and to provide, for multiple users, data and information for exploration of new mineral deposits, natural fertility for agriculture, and sources of natural and anthropogenic contamination that are considered harmful to human health [8]. As part of the PGAGEM, CPRM is cooperating with the Institute of Radiation Protection and Dosimetry (IRD) of the Brazilian National Commission on Nuclear Energy (CNEN) to develop the project Mapping the Radioactivity in Brazilian Soils (MAPRAD). Soil samples (0–20 cm depth) are collected with a stainless steel auger or paddles and the samples are labeled. A portion of the sample is sent for determination of chemical and physico-chemical properties. Another part, about 1 kg, is sent to IRD, where the samples are dried for 48 h in an oven, milled, sieved at 2 mm mesh, mixed, and stored in plastic bottles of 300 g capacity. Prior to analysis, the selected samples need to be stored for at least 30 d to achieve radioactive equilibrium between ^{226}Ra and ^{222}Rn. The samples are then sent to the laboratory for radionuclide activity concentration measurements by gamma spectrometry using a high purity germanium detector. The radium isotopes are determined by an indirect method: ^{226}Ra by averaging the ^{214}Pb and ^{214}Bi concentrations and ^{228}Ra by the concentration of ^{228}Ac. The determination of ^{40}K and ^{137}Cs is performed directly by their gamma peaks of 1460.8 and 661.6 keV, respectively. Owing to the large number of samples (about 10 000 samples are expected to be collected), initially only 30% will be analysed by gamma spectrometry. However, all samples will be processed, identified by sequential numbers, labeled and stored in a specially prepared soil storage room. The following strategy for ensuring the traceability of the samples and the quality of the analytical data was adopted in the MAPRAD project:

(a) All the information on geographical coordinates forwarded by CPRM is entered in the database system.
(b) The samples to be analysed are chosen based on a preview of the sample location in Brazil in accordance with the geographic coordinates provided by the staff of CPRM. The display is provided by Google Earth software. Figure 1 shows the locations of the samples collected to date.
(c) The control of the analytical quality is performed with two standard reference materials of soil that are analysed as blind samples within a set of 30 samples. The laboratory results are evaluated and entered in the web-based database.
(d) Samples for which the results are considered to be anomalous are reanalysed by another laboratory in order to compare the results.

FIG. 1. Sample locations.

3. RESULTS AND DISCUSSION

Since the beginning of the project, IRD has received 2882 soil samples, of which 705 (almost 25%) have been analysed so far. Up to now, samples from Brazilian states in the south-eastern region (São Paulo, Rio de Janeiro, Espírito Santo and Minas Gerais), the northern region (Pará and Roraima), the centre-west region (Mato Grosso do Sul and Goiás) and the north-eastern region (Pernambuco, Ceará, Paraíba, Alagoas and Bahia) have been collected. Except for the states of Bahia and Ceará, all data have been entered into the geographic information system (Fig. 1). However, for most of the states the sampling has not yet been accomplished. Sampling has been completed for Rio de Janeiro, Espírito Santo, São Paulo, Pernambuco, Paraíba and Alagoas states. A summary of the collected samples is shown in Table 1.

As the required time for each sample to be analysed is 60 000 s (16.67 h) and the IRD gamma spectrometry laboratories are not exclusively dedicated to this project, a completion date for the collected samples has not yet been established. Also, samples from other Brazilian states will be collected. So far, the concentration values obtained are in the ranges 3.1–174 Bq/kg for ^{226}Ra, 2.5–333 Bq/kg for ^{228}Ra and 2.4–2377 Bq/kg for ^{40}K. In most of the soils, the ^{137}Cs concentration is below the minimum detectable activity concentration of about 0.5 Bq/kg.

The results of are being used to generate scientific information on the occurrence of the radioelements in the Brazilian soil. So far, two doctoral studies are using the project results to establish reference values for radionuclides, three papers for presentation at conferences have already been written, and two papers are being published in scientific journals.

TABLE 1. SUMMARY OF SAMPLES COLLECTED IN THE MAPRAD PROJECT UP TO 14 SEPTEMBER 2016.

State	Received	Ready for analysis	Discarded	Analysed
Minas Gerais	704	209	8	189
São Paulo and Mato Grosso do Sul	729	187	42	164
Rio de Janeiro	56	0	2	54
Espírito Santo	66	6	1	59
Pará	307	112	10	66
Roraima	128	59	12	0
Pernambuco (with Fernando de Noronha)	183	0	6	173
Ceará	198	0	0	0
Alagoas	41	41	0	0
Paraíba	71	0	0	0
Bahia	185	0	0	0
Goiás	214	0	0	0
TOTALS	2882	614	81	705

4. CONCLUSIONS

The MAPRAD project contributes to the provision of public information on levels of radioactivity in Brazilian soil. A methodology to ensure the traceability of the soil samples has been established. A soil collection room has been established for these samples. An analytical quality control protocol has been established, which is verifying the quality of data from the three different laboratories. A system for providing data on the web has been developed and implemented. Samples from 13 Brazilian states are stored. The analytical results generated so far have been entered in the web-based system, and will become available to the public following publication in specialized technical journals, scientific theses and conference proceedings. A map of radioactivity in Brazilian soil is, for the first time, now being compiled.

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Poster Presentation

COMPARISON OF ^{210}Pb AND ^{210}Po ACTIVITY CONCENTRATIONS FOR SEDIMENT DATING

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Abstract

Records stored in natural archives, such as those for lake sediments, are used in environmental programmes for the assessment of changing erosion rates in a catchment arising from disturbances, and to monitor pollution by heavy metals and other contaminants. Accurate sediment chronologies are important to interpret those practices. One of the most important methods for dating recent sediments is through ^{210}Pb. This radionuclide occurs naturally as one of the ^{238}U decay series. It is widely distributed on Earth owing to its decay from radium in the ground or from radon that emanates to the atmosphere. The ^{210}Pb is deposited as particulates and falls into lakes where it is scavenged from the water column and deposited in the basins. The elevated ^{210}Pb concentrations are measured using the gamma spectrometry technique. However, this technique has a low sensitivity and small values are difficult to detect or require a long measurement time. The determination of ^{210}Po (a decay product of ^{210}Pb) using alpha spectrometry is more sensitive and rapid. The aim of this paper is to compare the activity concentrations of the two radionuclides in lake sediment samples to evaluate the use of ^{210}Po in the dating of sediments with low levels of ^{210}Pb.

1. INTRODUCTION

The increasing use and extraction of soil has been disturbing the natural environment and adding contaminants to water sources. The sediments deposited in lakes usually reflect the erosion processes in upstream catchments [1]. These sediments, previously considered only as nutrient sites, are now sources of contaminant information about water sources located near mining and ore processing industries and agricultural activities. Lake sediments stored in natural archives are used as records in a wide range of environmental programmes for purposes such as the assessment of changing erosion rates in a catchment (arising from disturbances such as afforestation, deforestation or changing agricultural practice) or the monitoring of pollution by heavy metals, organic pollutants and other contaminants [2]. Sediment quality is an important parameter in the assessment, protection and management of aquatic ecosystems. Because sediments influence the fate of many chemicals, concern exists about the potential impact on organisms that are exposed to sediments with elevated chemical concentrations [3].

One of the most important means for dating recent sediments (0–150 years) is through ^{210}Pb, which occurs naturally as one of the radionuclides in the ^{238}U series. Disequilibrium between ^{210}Pb and its parent isotope in the series, ^{226}Ra, arises through diffusion of the intermediate gaseous isotope ^{222}Rn. Some of the ^{222}Rn atoms produced by the decay of ^{226}Ra in soils escape into the atmosphere where they decay through a series of short-lived radionuclides to ^{210}Pb. This is removed from the atmosphere by precipitation or dry deposition, falling onto the land surface or into lakes and oceans. The ^{210}Pb falling directly into lakes is scavenged from the water column and deposited on the bed of the lake with the sediments [2]. The excess ^{210}Pb in the sediments, beyond the amount that is in equilibrium with the in situ ^{226}Ra, decays in accordance with Eq. (1).

$$C_{\mathrm{Pb}} = C_{\mathrm{Pb}}(0)\, e^{-\lambda t} + C_{\mathrm{Ra}}\left(1 - e^{-\lambda t}\right) \tag{1}$$

where C_{Pb} is the ^{210}Pb activity concentration in the sediment layer, $C_{\mathrm{Pb}}(0)$ is the initial ^{210}Pb activity concentration in the sediment layer at its time of formation, C_{Ra} is the ^{226}Ra activity concentration, λ is the ^{210}Pb decay constant and t is the time since sediment formation. This equation can be used to date a sediment provided a reliable estimate can be made of the initial ^{210}Pb activity $C_{\mathrm{Pb}}(0)$. Modelling and quantifying the process by which excess ^{210}Pb is produced and redeposited on the Earth's surface is an important prerequisite for the development of reliable methods for calculating ^{210}Pb dates [2]. In most cases the choice of method for determining ^{210}Pb activity concentration will be governed by what is available:

(1) Alpha spectrometry is more sensitive and most suitable for small samples of very low activity. The method is based on the measurement of ^{210}Pb via alpha radiation emitted by ^{210}Po, a decay product of ^{210}Pb. The ^{210}Pb is extracted from the sample by chemical digestion and deposited onto silver planchets for assay in a low background alpha spectrometer [2]. The detectors are simple and inexpensive, but it is necessary to have access to radiochemical facilities. A significant disadvantage is the time required to establish equilibrium between ^{210}Pb and ^{210}Po, or to allow the ingrowth of ^{210}Po. Furthermore, the method only determines total ^{210}Pb and ^{226}Ra concentrations [4].

(2) The gamma spectrometry method has a great advantage in that it does not require the leaching and radiochemical separation of ^{210}Pb or ^{210}Po [5]. A gamma assay requires minimal sample preparation, since gamma photons can travel significant distances without absorption. Furthermore, the measurements can be carried out on dried sediment samples without the need for radiochemical separation. The measurements are non-destructive so that, after gamma assay, samples can be used for further analyses. In addition, the method allows simultaneous determination of a range of radionuclides, including ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am. On the other hand, the method presents higher overheads and lower sensitivity for some radionuclides, and the efficiency calibration is particularly demanding at low energies [2].

2. METHODS

Four sediment samples (Bortolan 12, Bortolan 15, Bortolan 22 and Fazenda 03) were selected for measurement of ^{210}Pb and ^{210}Po activity concentrations. To ensure the quality and accuracy of the methods used, samples of certified reference material IAEA–447 (moss soil) were prepared as described in the IAEA reference sheet and analysed, one sample for lead and two for polonium. The analysis methods used for the sediment samples and the reference material were as follows:

(1) For ^{210}Pb determination, the sediment samples and reference material were dried at 110 ± 5°C for 24 h, and then removed and cooled in a desiccator for 1 h. A known mass of sample was homogenized and compressed into a 49×13 mm propylene container until full, and hermetically sealed with chloroform. Thirty days were allowed for ^{222}Rn ingrowth inside the container and for achieving equilibrium between ^{210}Pb and ^{226}Ra. The sample was then counted in a Canberra gamma spectrometry system with a high purity germanium detector of 45% relative efficiency until at least 1000 counts were reached. In order to create the efficiency curve, the efficiency was thoroughly set and then the samples and the reference material were evaluated.

(2) For ^{210}Po determination, the sediment samples were prepared using about 0.2 g of dried sediment and 1 mL of ^{209}Po tracer. The reference material was prepared using about 1 g of dried sediment and 10 mL of ^{209}Po tracer. The samples were transferred to a Teflon beaker and kept under agitation for 20 min for homogenization. Each sample was totally digested in an open system with mineral acids, and then the residue was dissolved using 1.5M HCl and filtered. Finally the solution was transferred to a deposition cell using 1.5M HCl and 1 g of ascorbic acid was added to eliminate interference. The polonium was deposited onto a silver planchet under agitation for 4 h at 80°C. Then, the planchet was counted on a surface barrier detector in an alpha spectrometry system.

3. RESULTS

For ^{210}Pb, the activity concentration in the reference sample was found to be 420 ± 30 Bq/kg, the same as the value specified in the reference sheet (420 ± 20) Bq/kg. The details are shown in Fig. 1. For ^{210}Po, the activity concentrations in the two reference samples were found to be 345 ± 15 and 330 ± 15 Bq/kg. The activity concentration specified in the reference sheet, after applying a decay correction for the current date, was 344 ± 8 Bq/kg. The energy peaks for the first of the reference samples are shown in Fig. 2.

Both measurement techniques were then applied to the four sediment samples. The results are shown in Table 1.

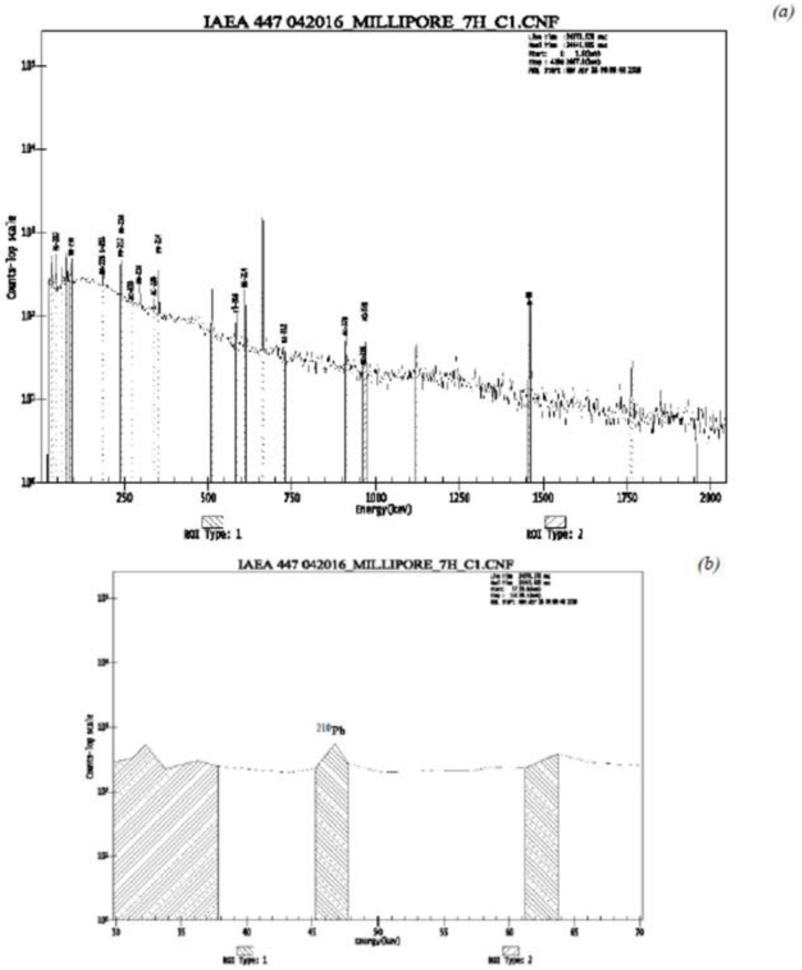

FIG. 1. IAEA–447 reference sample: (a) Elements identified by gamma spectrometry; (b) Detail of lead spectrum.

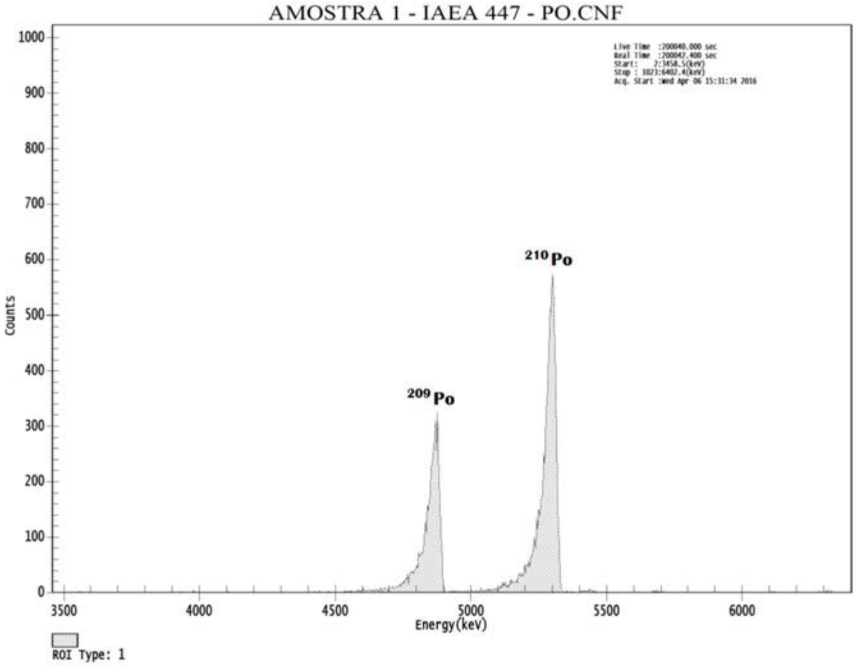

FIG. 2. IAEA–447 reference sample, alpha energy peaks for ^{210}Po and ^{209}Po.

TABLE 1. ACTIVITY CONCENTRATIONS MEASURED IN SEDIMENT SAMPLES.

	Activity concentration (Bq/kg)	
	Pb-210	Po-210
Sample 1	294.1 ± 20.9	258.9 ± 17.9
Sample 2	279.6 ± 19.4	246.0 ±17.2
Sample 3	289.8 ± 11.8	257.5 ± 18.3
Sample 4	301.1 ± 19.2	299.2 ± 16.2

4. DISCUSSION

The results obtained through gamma spectrometry for ^{210}Pb are slightly higher than those obtained through alpha spectrometry for ^{210}Po. This difference could be explained by losses in the radiochemical analysis or the amount of tracer added to the sample, that could have an activity concentration closer to the one found. The results were considered to be satisfactory for both methods. When the ^{210}Pb levels in the collected samples are high and there is a suitable amount of material available, it would be appropriate to choose the gamma spectrometry technique for performing further analyses. While the gamma spectrometry technique requires a longer time, to allow for the ingrowth of ^{222}Rn inside the flask, it involves less sample manipulation.

5. CONCLUSION

The values obtained using the gamma spectrometry method for ^{210}Pb analysis and using the radiochemical method for ^{210}Po analysis were considered satisfactory, showing that either method can be used. These results corroborate the intrinsic relationship of radioactive equilibrium between ^{210}Pb and its progeny ^{210}Po, proving the existence of a chemical balance as proposed in Ref. [5]. Since there is a substantial amount of ^{210}Pb in the analysed sediment samples, the gamma spectrometry technique will be used for further determinations of ^{210}Pb activity concentration and subsequent sediment dating.

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ENVIRONMENTAL IMPACT ASSOCIATED WITH NORM INDUSTRIES

(Session 5)

Chairperson

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RADIOLOGICAL EVALUATION OF MONAZITE MINING IN CENTRAL SPAIN

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Abstract

The Mulas rare earth mining project at Ciudad Real in central Spain, 200 km south of Madrid, is associated with an unconsolidated sandy and clayey alluvium–colluvium deposit originating from paleozoic weathered source rocks that host 0.1–2.0 mm diameter monazite nodules containing about 60 wt% rare earth oxides. This ‘grey’ or ‘dark’ monazite is completely different from the common ‘yellow’ thorium enriched monazite that was mined in the recent past as a source of rare earths. A detailed radiological evaluation has been performed associated with the mining of the Mulas monazite. This evaluation was based on the following studies: (a) determination of the activity concentrations of several radionuclides from the uranium and thorium series in representative samples of the material mined, (b) construction of an external gamma dose rate map of the mining area, (c) study of the distribution of the natural radioactivity in the material extracted as a function of the grain size, (d) radon determinations in the area, and (e) laboratory leaching experiments. Although rare earth mining is one of the activities recognized in the ‘positive list’ of NORM activities, all the results indicate that this rare earth mining activity performed in central Spain can be considered as being outside the scope of international radiation protection standards.

1. INTRODUCTION

The grey monazite deposit called ‘Matamulas’, located in central Spain, 200 km south of Madrid, is the first rare earth deposit to be mined in the country. It was discovered in 1989, and consists of monazite dispersed in an alluvial–colluvium deposit in the form of nodules of sizes between 0.1 and 2 mm. The monazite to be mined is particularly enriched in Nd and Eu. The mining site covers an area of about 500 ha and contains superficial monazite deposits (within 100–200 cm of the surface) in concentrations of 2.5–3.0 kg/m^3. For that reason, opencast mining is performed using conventional digging equipment. The ‘grey’ or ‘dark’ monazite, containing about 60 wt% rare earth oxides, 23 wt% P_2O_5, 10 wt% SiO_2 and 7 wt% iron oxides, is a rare earth phosphate mineral of diagenetic–metamorphic origin and high density (4650 kg/m^3). It is completely different from the common ‘yellow’ thorium enriched monazite that was mined in the recent past as a source of rare earths. The monazite is separated from the host material by applying grain size and other physical separation techniques that exploit the presence of nodules and the high density.

2. ACTIVITY CONCENTRATIONS

The radiological study began by collecting six representative samples of the mined material, taken from different zones of the mining area. From these samples, rock particles greater than 1 cm in size were removed and the remaining material was subjected to radiometric analysis using high resolution gamma-ray spectrometry with a Canberra low energy germanium detector shielded with 10 cm of lead. The homogenized samples were placed in Petri dishes and the activity concentrations of the different radionuclides were determined after the performance of an efficiency calibration corrected for self-absorption and coincidence effects through the application of the LABSOCS code based on a Monte Carlo simulation. This non-destructive gamma spectrometric technique enabled the activity concentrations of the following radionuclides of natural origin to be measured: ^{7}Be, ^{40}K, ^{208}Tl, ^{210}Pb, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{228}Ac and ^{234}Th. All the uncertainties were calculated with $k = 1$. Results obtained from the gamma ray spectrometric measurements are shown in Table 1.

TABLE 1. RADIONUCLIDE CONCENTRATIONS IN SAMPLES COLLECTED FROM THE MATAMULAS DEPOSIT

	Activity concentration (Bq/kg)					
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6
Th-232	94 ± 12	82 ± 10	91 ± 4	77 ± 5	105 ± 6	66 ± 4
Th-230	60 ± 3	55 ± 3	47 ± 4	43 ± 2	50 ± 3	46 ± 3
U-238	52 ± 12	51 ± 12	46 ± 10	51 ± 10	57 ± 12	42 ± 10
U-234	47 ± 3	45 ± 3	49 ± 3	44 ± 4	54 ± 3	49 ± 2
K-40	740 ± 41	594 ± 37	851 ± 39	824 ± 43	688 ± 32	845 ± 40
Cs-137	2.3 ± 0.3	<1.0	<1.0	3.1 ± 0.4	3.0 ± 1.0	<1.0

The following comments can be made on the results in Table 1:

(a) Results are shown only for the most representative radionuclides, and therefore exclude most of the uranium and thorium series radionuclides for which secular equilibrium conditions can be expected in the undisturbed samples that were not affected by anthropogenic activities prior to their collection.

(b) Some of the samples contained traces of ^{137}Cs originating from fallout from the atmospheric nuclear tests performed in the 1950s and 1960s — this demonstrated the sensitivity of the radiometric technique and reflected the superficial location of the samples.

(c) The results indicated uniform activity concentrations of uranium and thorium series radionuclides across the mining area (Th series: 70–100 Bq/kg, U series: 40–60 Bq/kg, ^{40}K: 600–850 Bq/kg), indicating that the number of samples analysed was sufficient to enable conclusions to be drawn about the radiological implications of this mining activity.

(d) The activity concentrations of ^{232}Th and ^{238}U are clearly below the criterion of 1000 Bq/kg specified in international standards, above which the material might be subject to regulation and therefore considered as NORM. The activity concentrations are similar to those found in soil in many areas of Spain, underlining the negligible radiological impact of this mining activity.

(e) The results are not regarded as surprising because, although the monazite is enriched in Th and U series radionuclides, the nodules in which it is contained are dispersed in the host material at concentrations of 2.5–3.0 kg/m^3. Determinations performed in a representative sample of monazite nodules extracted from the deposit indicated activity concentrations of 8000 Bq/kg for ^{232}Th and 2000 Bq/kg for ^{238}U.

(f) Atmospheric radon concentrations in the mining area were found to be similar to normal background values. This finding is consistent with the low ^{238}U activity concentrations, the existence of secular equilibrium in the ^{238}U series and the opencast method of mining. The inhalation of radon and its progeny is therefore of no radiological concern.

3. GAMMA DOSE RATES

The results and conclusions obtained from the gamma-ray spectrometric determinations were confirmed through the performance of a set of external gamma dose rate measurements across the deposit. These measurements were carried out using a Berthold UMo LB 171 radiation monitor, calibrated periodically and sensitive in an appropriate energy range from 30 keV to more than 2 MeV. This monitor works in the proportional mode, is specially designed for the measurement of low dose rates and has a detection limit of 0.055 μSv/h. The measurements were carried out at each selected location for 120 s in order to achieve measurement uncertainties below 10%. The results are shown in Fig. 1. The dose rates were quite uniform across the deposit, consistent with the results of the radiometric determinations. The gamma dose rate values are a factor of 2 higher than the representative background values for Spain. This correlates well with the finding that the Th and U concentrations were 2–3 times higher than those in normal soils throughout the country. The theoretical increment in external gamma dose received by the workers carrying out the mining activities would be, at most, 0.10–0.15 μSv/h, leading to an incremental occupational effective dose of 0.2–0.3 mSv/a (assuming 2000 h per year of presence of the affected workers in the mining area). This maximum increment in effective dose is clearly lower than the exemption level of 1 mSv/a in international standards, confirming again that there is no need for radiation protection measures.

4. DISTRIBUTION OF RADIOACTIVITY WITHIN THE MINED MATERIAL

Once the raw material is extracted from the mine, it is transported to the production plant, where the monazite nodules are separated by means of grain size and physical separation processes shown schematically in Fig. 2. The raw material is divided into three size fractions, with the monazite being concentrated in the intermediate fraction.

Representative aliquots of each of the three fractions from the separation process were analysed by high resolution gamma ray spectrometry. The results are shown in Fig. 3. The following comments can be made:

(a) The distribution of U and Th between the analysed fractions is inhomogeneous. The fine and coarse fractions have U and Th activity concentrations well below the exemption levels specified in international standards, while the concentrations in the intermediate (monazite rich) fraction are significantly higher. The monazite is not only enriched in rare earths, but also in Th and U.

(b) Due to the low U and Th content in the fine and coarse fractions, both can be considered as non-radioactive material with no radiological implications associated with their storage or transport. These fractions, representing 80% of the raw material initially mined, can be returned to the mining area for permanent disposal without the need for radiation protection measures.

(c) The fine fraction, characterized by a low U and Th content, can be resuspended into the air and inhaled by workers and/or members of the public. The fraction enriched in monazite, and thus in Th and U, has a density and a grain size that prevents its resuspension and possible inhalation. Consequently, the radiological impact through the inhalation route associated with the mining activity can be considered negligible.

Observation of the monazite nodules present in the intermediate fraction using scanning electron microscopy confirmed that the enriched concentrations of Th and U were encapsulated in the monazite nodules, preventing any possible migration. Preliminary leaching experiments applied to the intermediate fraction, using slightly acidified water, confirmed this conclusion because no detectable U and Th levels were found in the leachate fractions.

FIG. 1. External gamma dose rates measured across the mining site.

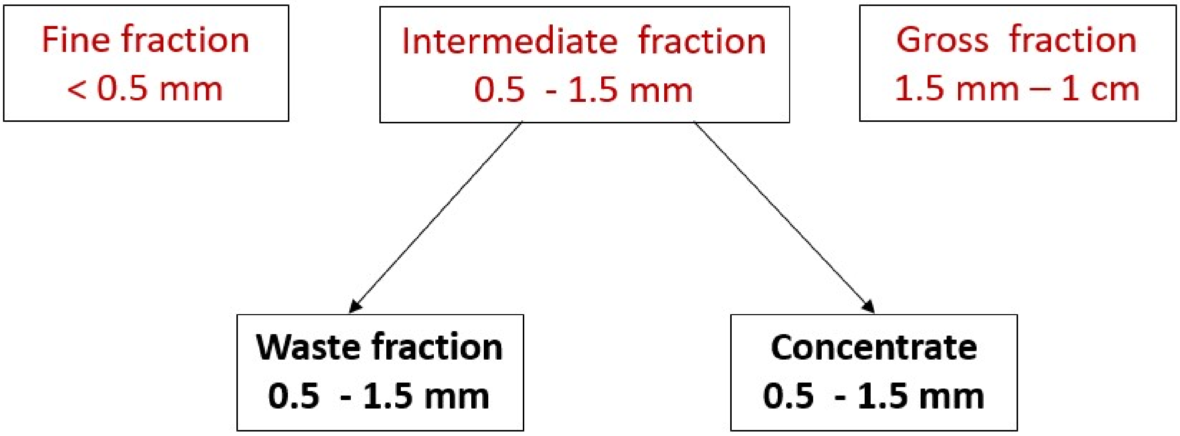

FIG. 2. Grain size fractionation applied to the raw material mined from the mineral deposit.

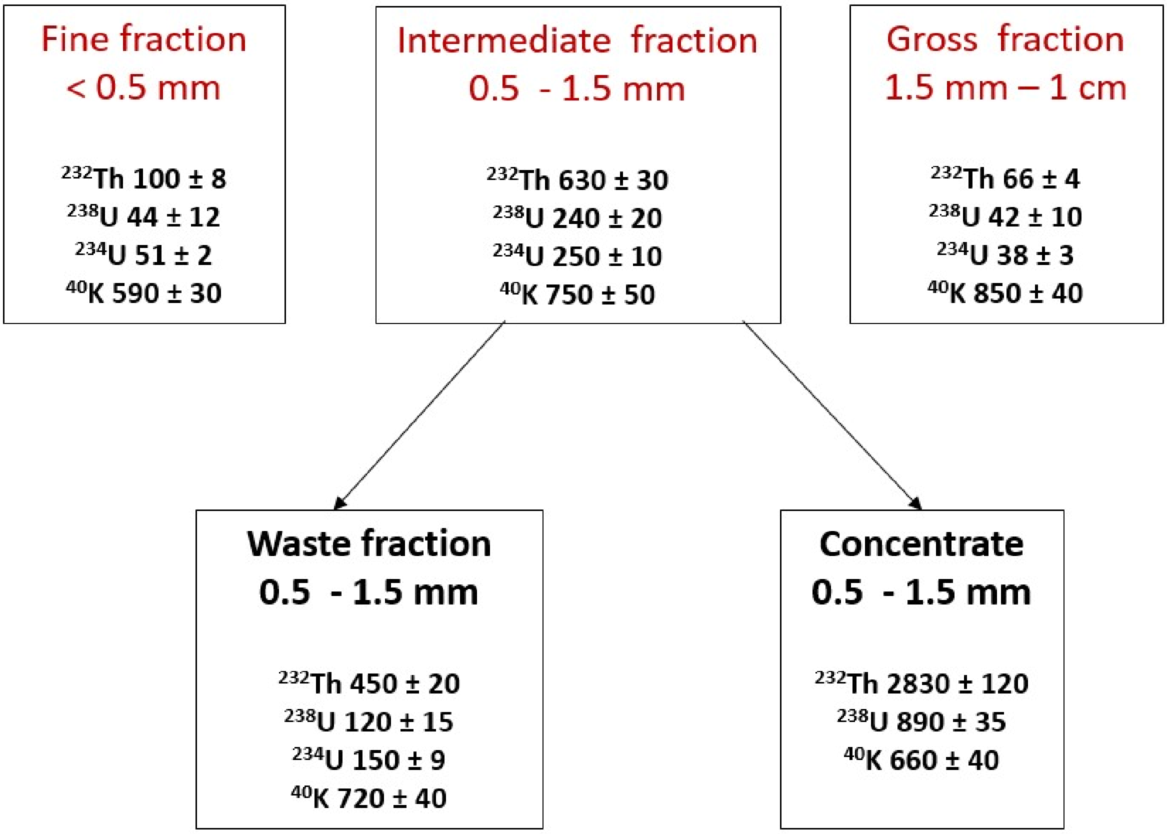

FIG. 3. Radionuclide activity concentrations in the three size fractions.

5. CONCLUSION

All the experiments and measurements indicate that the occupational and public radiological impact arising from the mining of the Matamulas monazite deposit is negligible, and no radiation protection measures need to be taken. However, this conclusion should not be extrapolated to the activities performed in the processing plant, which need an independent radiological evaluation.

ENVIRONMENTAL CONTAMINATION AROUND THE CALDAS URANIUM MINE DUE TO URANIUM AND THORIUM IN SEDIMENTS, ROCKS, MINE WASTE AND FERTILIZER

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Abstract

The Osamu Utsumi uranium mine is located on the alkaline plateau complex of Poços de Caldas, MG, Brazil, where there are several radioactive anomalies. This facility has been undergoing a decommissioning process by Brazilian nuclear and environmental regulatory agencies In order to meet the technical and legal requirements for mine closure, the characterization of the existing environment and proposals for specific interventions are required. The area surrounding the mine contains fields and pastures, vegetable cultivation, a commercial rosarium, native forests and, nowadays, mainly commercial forestry. These are plantations that use significant amounts of fertilizer, which can eventually contribute to environmental contamination. In order to determine the levels of uranium and thorium contamination resulting from these activities, a research project was carried out in the area. This specific part of the study involved two steps: (i) the collection of bottom sediments from local rivers, unweathered rocks, mining waste and fertilizer; and (ii) the measurement of uranium and thorium levels by neutron activation analysis. The results demonstrated that the sediments collected close to the mine presented higher uranium concentrations, showing the influence of the uranium mine in the area. On the other hand, no conclusive results were found concerning potential contamination caused by anthropogenic or natural sources of thorium.

1. INTRODUCTION

Environmental contamination began when men started using agriculture to ensure the survival of the human race. Initially, this relationship was a balanced one. Men were only planters and hunters, living in an integrated manner with the ecosystem. However, as time went by, agriculture took on new proportions. More than just the basic agricultural activities became necessary to guarantee subsistence and then humans started interfering with the environmental equilibrium.

Mineral exploration became a major activity during the first industrial revolution, as the demands of mass production created an intensification of mineral extraction to supply the increasing industrial activities. In practice, mining activities have become synonymous with suppression of natural vegetation or the hindering of its regeneration [1]. The removal of high fertility superficial soil causes, in the remaining exposed soils, possible physical and chemical effects where the water downstream of the open facility is affected by turbidity due to suspended fine sediments and by silting resulting from erosion.

Phosphate rock is the main type of mineral used in the production of phosphate fertilizer. This can contain elevated concentrations of radionuclides of natural origin. The preoccupation concerning environmental contamination is related to the fertilizer production process, where most of the metals from the phosphate rock remain within the product [2, 3]. In addition, the concentrations of radioactive lead, uranium and thorium isotopes, as well as their decay progeny, can be significant in fertilizer products [4]. The study area was located on the Poços de Caldas plateau, in the municipality of Poços de Caldas, MG, covering an area of 102 km^2, including part of the mining treatment unit (UTM-Caldas) of Indústrias Nucleares do Brasil (INB). The Poços de Caldas mining and industrial complex has contributed greatly to the Brazilian nuclear programme and, more specifically, to the development of the nuclear fuel cycle for the generation of electric power. At present, the facility is undergoing a decommissioning process that is being carried out by the National Nuclear Energy Commission (CNEN) and by the Brazilian Environmental Institute (IBAMA) [5]. In addition to UTM-Caldas, the interest area has fields and pastures, vegetable cultivation, a commercial rosarium, native forest and, nowadays, mainly commercial forestry. It is important to mention that, in general, these types of cultivation, especially forestry, use significant amounts of phosphate fertilizer and soil amendments. This paper describes the investigation of environmental contamination levels through the determination of uranium and thorium concentrations in samples of fertilizer, bottom sediments from local rivers, mining waste and unweathered rocks collected in the study area.

2. METHODS

The study area includes a vast hydrographical network in the Poços de Caldas plateau region, encompassing three sub-catchments of the Pardo river catchment. Through the data analysis done and based on soil use and occupation in the region, along with ease of access considerations, sediment sampling points were selected. Twenty nine sediment samples were collected from the beds of the Consulta Creek, Soberbo Stream and Taquari River, the main receiving water bodies for liquid effluents in the study area. In addition, ten mining waste samples, three unweathered or slightly weathered rock samples and seven types of fertilizer used by farmers in the region were collected. Figure 1 shows the location of the sediment and mining waste sampling points, as well as the radioactive anomalies found in the region. The determination of uranium and thorium concentrations in the samples was carried out by means of neutron activation analysis using the methods of retarded neutrons and k0-AAN, respectively, at the Nuclear Energy Development Centre (CDTN) [6].

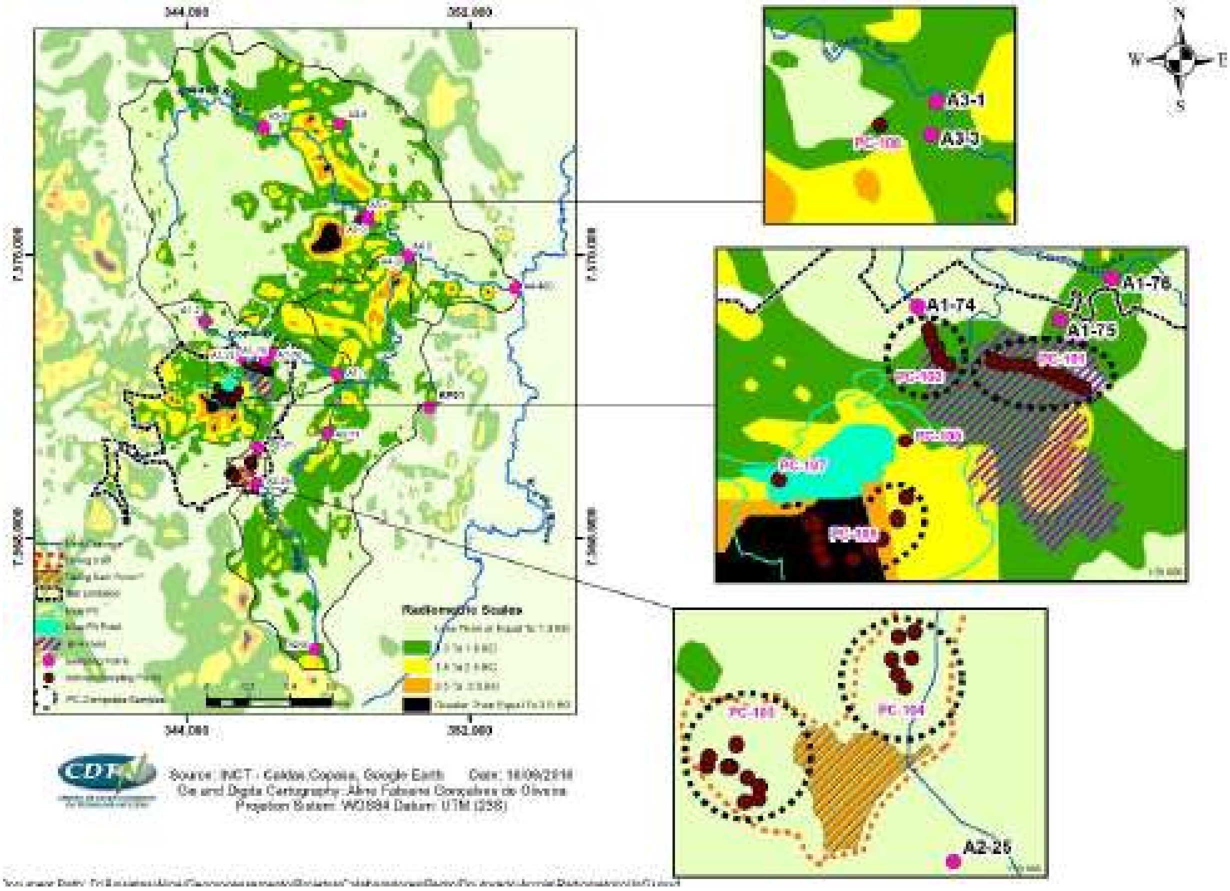

FIG. 1. Locations of the sampling points and the radioactive anomalies [6].

3. RESULTS AND DISCUSSION

The uranium and thorium concentration results are presented in Figs 2–5 and Figs 6–9, respectively. Analysis of the results verified the presence of elevated uranium concentrations in one of the mining waste samples (SCEXT08), which is a gneiss originating from the mining waste treatment process.

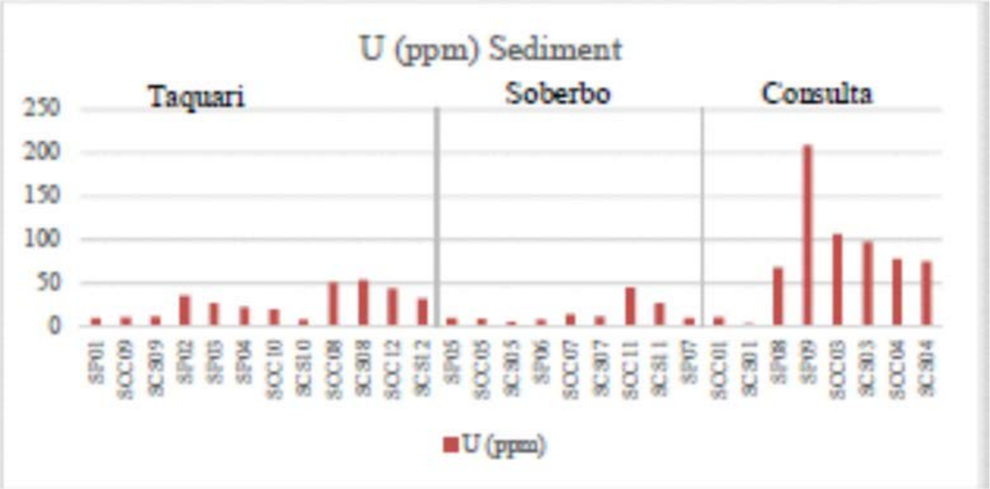

FIG. 2. Uranium concentrations in sediments.

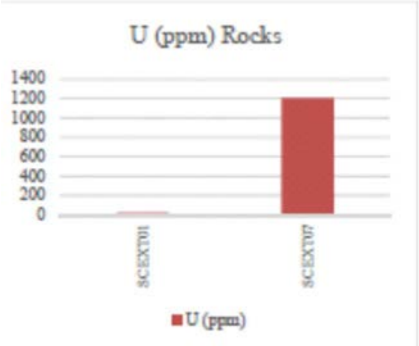

FIG. 3. Uranium concentrations in rocks.

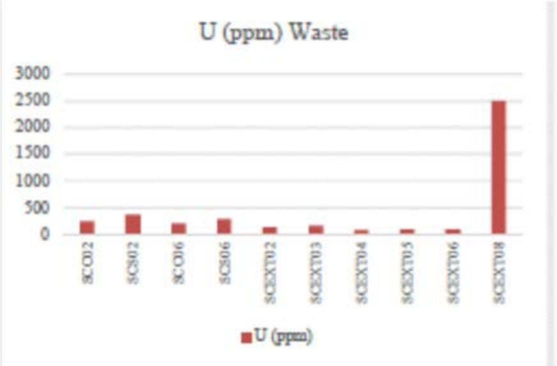

FIG. 4. Uranium concentrations in mining waste.

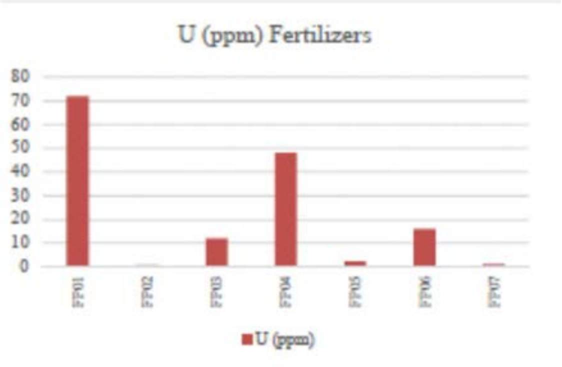

FIG. 5. Uranium concentrations in fertilizer.

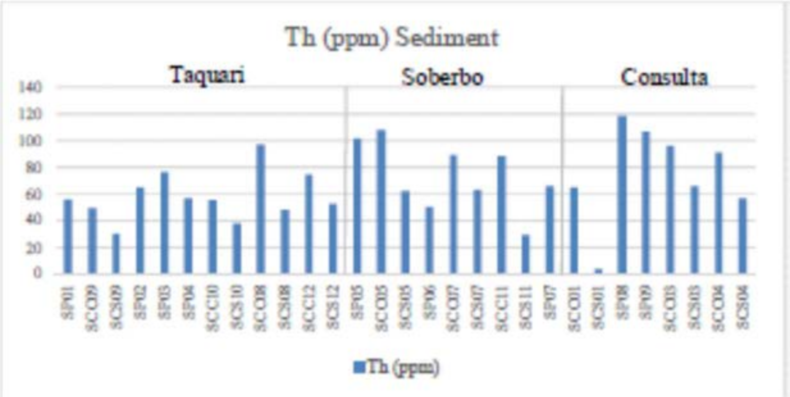

FIG. 6. Thorium concentrations in sediments.

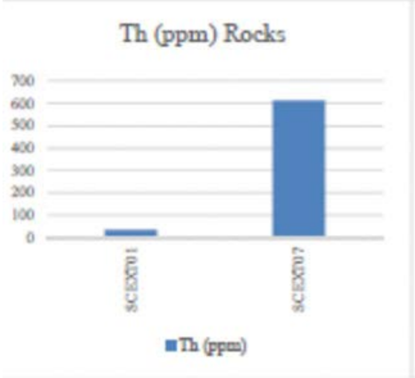

FIG. 7. Thorium concentrations in rocks.

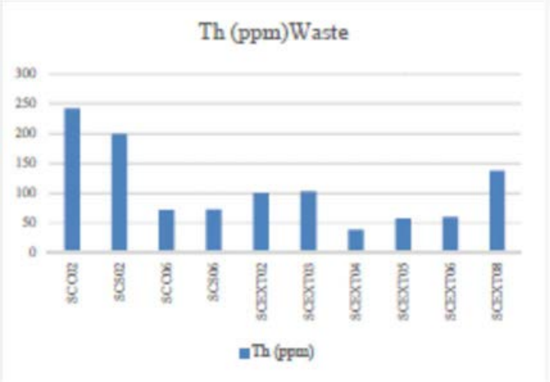

FIG. 8. Thorium concentrations in mining waste.

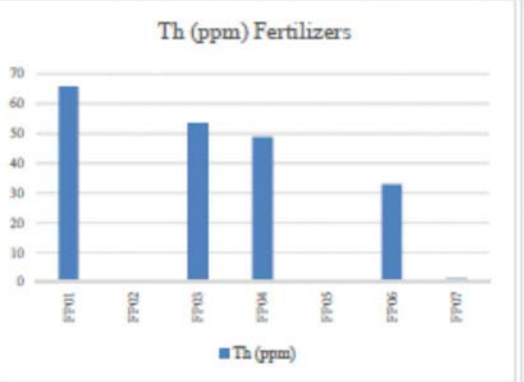

FIG. 9. Thorium concentrations in fertilizer.

In sediments, higher uranium concentrations were found in the samples collected from the right bank of the Consulta Creek (sample SP09) and more specifically within the UTM-Caldas facility. This result may be explained by the geological anomaly that gave rise to the opening of the first uranium mine in Brazil, or even by the uranium mining and extraction operation itself. The other points (SCC03, SCS03, SCC04 and CSS04), located just downstream of the effluent discharge point of the Consulta Creek, presented lower values. Nevertheless, they were much higher than the values found in other water bodies that did not suffer direct influence from the mine. In Figs 6–9, it is evident that the mining waste samples also presented high concentrations of thorium. The samples collected in the Nestor Figueiredo catchment (SCS02/SCC02) were the ones that showed higher concentrations. It is worth noting that this basin was constructed in order to retain water that came from the mine waste pile. Uranium and thorium activity concentrations in mining waste and fertilizer samples were also determined in order to correlate them with the values found in the sediment samples. However, through this specific analysis, a definitive statement concerning thorium behaviour was not possible, since this element, unlike uranium, exhibits practically no movement in aqueous environments.

4. CONCLUSION

The results indicate the presence of elevated uranium concentrations in the samples of sediments collected in the basin of the Consulta Creek, especially within the mine facility and just downstream of the effluent discharge points, demonstrating the influence of mining activities. Conclusive results were not found concerning the possible contamination potential caused by anthropogenic or natural thorium sources. Additional studies, using techniques for the determination of the isotopic ratio Pb/Po, have been performed in order to obtain a more detailed knowledge of the geochemical behaviour of these elements and of the source of contamination in the area.

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RADIATION EXPOSURES FROM LANDFILL DISPOSAL OF TITANIUM DIOXIDE INDUSTRY WASTE: COMPARISON OF MODELS WITH SITE SPECIFIC MEASUREMENTS

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Abstract

Large quantities of calcium based reaction residue are produced by a titanium dioxide production plant. There are currently no options for the use or recycling of this residue, and instead it is disposed of as waste to a non-hazardous waste landfill. The waste contains radionuclides from the ^{232}Th decay series and, to a lesser extent, from the ^{238}U decay series, with a total activity concentration in the range 1–3 Bq/g. This paper discusses the regulatory regime in the United Kingdom relevant to the disposal of 'exempt NORM waste' (up to 10 Bq/g) without the requirement for radioactive waste disposal licensing. Estimations of occupational and public radiation exposures made using environmental models are also included. Re-evaluation of occupational doses (i.e. to landfill workers) made using on-site measurements of gamma dose rate and individual doses (using electronic and passive personal dosimeters) are also presented. The results obtained using monitoring data are compared with those obtained from environmental modelling, and the importance of using site specific monitoring data, where available, is discussed.

1. INTRODUCTION AND REGULATORY FRAMEWORK

More than 100 000 t/a of calcium based reaction residue is produced by a titanium dioxide production plant. The residue contains radionuclides of natural origin, predominantly from the ^{232}Th decay series, with a smaller contribution from the ^{238}U decay series. The residue has been routinely analysed by gamma spectrometry for over 10 years and was found to have a total activity concentration (in terms of the sum of the concentrations of the single radionuclides with the highest activity concentrations in each of the ^{238}U and ^{232}Th decay chains) in the range 1–3 Bq/g. There are currently no options for the use or recycling of this residue, and instead it is disposed of as waste at a non-hazardous waste landfill site. The principal legislation for regulating radioactive substances is the Environmental Permitting (England and Wales) (Amendment) Regulations 2011 [1] in England and Wales and the Radioactive Substances Act 1993 [2] in Scotland and Northern Ireland. These also contain provisions for the exemption of disposal of wastes containing radionuclides of natural origin from the Regulations [1, 3, 4]. The concept of exemption is applied through a two tier process:

(1) 'Out of scope': The waste[1] is not defined as radioactive and is therefore unconditionally exempted from the requirements of the regulations. Only waste arising from a defined list of NORM industries (which includes manufacture of titanium dioxide pigments) and which exceeds specified activity concentrations (e.g. 0.5 Bq/g for ^{238}U and ^{232}Th assumed to be in secular equilibrium with their progeny) are considered to be in scope.

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[1] For NORM industries in which the radioactivity is incidental, only waste is potentially in scope. Raw materials and intermediates are not deemed radioactive for the purposes of licensing.

(2) 'Exempt NORM waste': A conditional exemption under which solid NORM waste that is in the scope of the legislation (i.e. defined as radioactive) may be disposed of without licensing. This waste is still subject to some regulatory requirements, such as record keeping.

There are two categories of exempt NORM waste called Type 1 and Type 2. Type 1 is based on a generic dose assessment [5] which demonstrates that annual doses to the public and landfill workers are below 0.3 mSv and 1 mSv, respectively, with an additional consideration of 3 mSv for inadvertent intrusion [6]. It allows for NORM waste containing less than 5 Bq/g to be exempted from licensing provided that the total amount of waste disposed of per year to landfill does not exceed 5×10^{10} Bq.

Type 2 NORM waste provides an additional *site specific* exemption where either the activity concentration is less than 10 Bq/g, or the annual activity disposed of exceeds 5×10^{10} Bq. This requires that a specific radiological assessment is carried out and approved by the regulatory body to demonstrate that the same annual dose criteria for the public and landfill workers are met. This paper describes the specific radiological assessment for disposal of Type 2 NORM waste from a plant manufacturing titanium dioxide pigments.

2. SCENARIOS CONSIDERED USING ENVIRONMENTAL MODELS

The environmental regulatory bodies produced a template which sets out the nature of the dose assessment and the level of detail required. It specifies 27 potential exposure scenarios for the public and workers during the operational and post closure phases of the site which are either likely to occur (e.g. workers handling waste, public exposure resulting from groundwater migration from a major or a minor aquifer under the site), or not certain to occur (e.g. fires, flooding and inadvertent intrusion). Not all the scenarios may be relevant to a specific site.

The site under consideration is located in an industrial area close to a saline estuary (about 250 m away at the closest point) with housing and agricultural areas being 2 and 5 km away respectively. The flow of the water table is towards the estuary and there is saline intrusion into the local aquifer; there are no public extraction points from the aquifer. The site has a clay liner at the base which prevents leachate from entering the sediments below; the leachate collection points are at the base of the landfill site. For the assessment, a radionuclide activity concentration of 1 Bq/g for each chain (^{238}U and ^{232}Th) in secular equilibrium with its progeny was assumed with 100 000 t/a of waste being co-disposed with 50 000 t/a of non-radioactive waste in a non-hazardous waste void.

Given that any exposure of the public will result from activity concentrations at levels which may be hard to detect above background levels and may not occur until many years into the future, the use of predictive models to assess doses is necessary. For members of the public, two scenarios were considered during the operational phase of the site: discharges of the leachate to the nearby estuary and flooding of the site. It was cautiously assumed that the discharge occurs directly into the estuary although, in reality, the leachate is treated in lined reed bed cells, with the effluent being subsequently discharged into the estuary. For the flooding scenario it was assumed that there was an overflow of leachate over the sides of the engineered barrier with the activity draining into the nearby river. Doses to the public were calculated using PC-CREAM 08 which is a software implementation of the European Union methodology [7] to assess the radiological impact of routine discharges of radioactive material into the environment.

For the assessment of doses to landfill workers, handling of the waste and exposure resulting from a fire were considered. The methodology for calculating doses received by landfill workers from handling waste is detailed in Ref. [8] with the external dose rates for the natural decay chains given in Ref. [5]. Doses for exposure from a fire at the site were based on the method given in Ref. [9], with workers assumed to be working 100 m downwind of the fire.

Fifty years after closure of the site, doses received by members of the public were calculated for the following scenarios: migration of the leachate into the nearby river, flooding and coastal erosion of the site and inadvertent intrusion due to housing being built on the site. For the flooding and inadvertent intrusion scenarios it was assumed that the site was restored to be used for housing and the doses were calculated using the methodology detailed in Ref. [10]. Doses to householders from radon emanating from the waste directly below the house were also estimated [9]. For coastal erosion it was conservatively assumed that the entire landfill is washed into the sea in a single year and the doses were calculated using PC-CREAM 08.

The highest dose calculated for the public was 1.3 mSv/a (65% of the dose being from inhalation of radon) for the inadvertent intrusion scenario where houses are assumed to be built on the site 50 years after closure. This dose is lower than the dose criterion for inadvertent intrusion of 3 mSv/a. Doses from the other scenarios are many orders of magnitude lower than this and considerably lower than the dose criterion of 0.3 mSv/a for members of the public.

For landfill workers, it was assumed they spend 2000 h/a at the site close to the uncovered waste. The external dose rates were calculated at 1 m above the surface of the waste using Microshield [11] for gamma radiation and the methodology given in Ref. [12] for beta radiation. Doses from inhalation of suspended dust were calculated by assuming that the activity concentration in the dust was at the same average activity concentration in the waste. Doses for the inhalation of leachate sprayed onto the surface of the landfill were cautiously estimated based on the workers standing in the spray for 700 h/a [13]. In terms of skin contamination it was assumed that no gloves are worn and that the backs of the workers' hands are covered with a thin layer of waste. Although workers periodically wash their hands, it is probable that waste attaches to their hands shortly after recommencing work and the calculation therefore cautiously assumed that the waste remained on the workers' hands for the whole of their shift. This was also assumed to lead to inadvertent ingestion.

Based on these assumptions an annual radiation dose for landfill workers of 1.9 mSv (over 85% due to external radiation) was calculated. This value exceeds the dose criterion of 1 mSv per year for landfill workers, and some site specific measurements were commissioned to investigate further.

3. SITE SPECIFIC MONITORING

Public Health England visited the site to make on-site measurements of gamma dose rates and individual external doses and to obtain information on working patterns. It was observed that the waste is delivered by lorry, loose tipped on the landfill and then layered and covered by a mechanical excavator and bulldozer: there is no direct handling of the waste. It was noted that the waste remains damp and sticks together, and therefore there is no potential for significant levels of airborne dust during the disposal process. Consequently, only exposures from external gamma radiation were considered.

Gamma dose rates varied from background to 1.5 μSv/h in close contact with bulk waste piles, with readings in the cabs of the excavator and bulldozer being 0.3–0.4 μSv/h. The disposal operation is continuous (2000 h/a), and this produces an effective dose in the range 0.6–0.8 mSv/a.

Electronic personal dosimeters were worn for a two week period by the operators of the excavator and bulldozer. The highest estimated dose was received by the excavator operator; the dose to the bulldozer operator being significantly lower. Based on the assumption that the excavator operator works on site for 250 days per year (i.e. 5 days per week, 50 weeks per year), the highest estimated dose is 0.6 mSv/a. This value is consistent with the dose estimated from the gamma dose rate measurements, which is to be expected, given the relatively constant working patterns.

In addition, the same two landfill workers were also issued with passive personal dosimeters (thermoluminescent dosimeters), with a 3 month wear period. The results from these dosimeters indicate an estimated dose of 0.4 mSv/a.

4. DISCUSSION AND CONCLUSIONS

The assessment of doses received by members of the public from the disposal of NORM waste to landfill inevitably have to be based on environmental modelling. In this case, all the estimated doses were well below the public dose constraints of 0.3 mSv/a for all scenarios except inadvertent intrusion where the dose criterion of 3 mSv/a applies.

The doses to landfill workers are also normally assessed by modelling; however, these estimates can be verified through on-site measurements as soon as the disposal process has started. The results presented in this paper demonstrate the value of such on-site measurements, especially where estimated doses are close to 1 mSv/a, which is the common national and international criterion for the application of regulatory control to NORM.

In the case considered in this paper, the exposure conditions for landfill workers were very consistent and predictable across the working year and therefore provide the best opportunity to estimate doses from on-site measurements consistent with the doses predicted from modelling. The results indicate that the doses to landfill workers predicted by modelling are overestimated by a factor of three to four when compared with those calculated using measured gamma dose rates.

Individual dosimeters are the most reliable means of assessing annual dose; they also usually give the lowest results (i.e. because they do not involve pessimistic assumptions), as seen in this case. Such dosimeters are not routinely issued to NORM workers, and care is needed to explain to workers the reason for their use. In this case, the use of individual dosimeters provided reassurance to workers and also direct evidence to the regulatory authority that the exemption criterion for NORM waste disposal was being met. For these reasons, the disposal company is intending to continue using individual dosimeters on a routine basis.

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RADIONUCLIDES OF NATURAL ORIGIN IN STREAM SEDIMENTS OF THE POÇOS DE CALDAS PLATEAU, BRAZIL

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Abstract

The Poços de Caldas plateau is well known for its anomalous natural radioactivity. This region hosts the first Brazilian uranium mine, which was established in the 1970s and operated from 1982 to 1995. This facility is currently under a closing process and an environmental remediation strategy is being formulated. Due to the acid mine drainage that occurs in this installation, a continuous treatment of approximately 350 m^3/h of water containing radionuclides of natural origin and other metals leached from the waste rock piles and mine pit has become necessary, incurring high financial costs every year. This installation discharges effluent into the Ribeirão das Antas River. Draining 70% of the area of the Poços de Caldas plateau, the river is of strategic importance to the city of Poços de Caldas (population approximately 150 000). The aim of this research was to determine whether the radioactivity found in sediments along the Ribeirão das Antas River (~60 km) is from natural or anthropogenic sources. Sediment samples were collected at 8 stations along the river course in 2015 and 2016. The determination covered U, Th, ^{226}Ra, ^{228}Ra and ^{210}Pb.

1. INTRODUCTION

The Poços de Caldas plateau is located in the south-west of the Federal State of Minas Gerais, Brazil (21°48'20.5"S, 46°34'22.9"W) and is well known for its unique geology [1]. It is a volcanic caldera about 1200 m above sea level with a circular shape approximately 35 km in diameter. The plateau is also known for its radioactivity anomalies associated with uranium and thorium deposits. The first Brazilian uranium mine is located in the area and operated from 1982 to 1995. Now, it is to be decommissioned and environmental remediation of the site is expected to be carried out. Acid mine drainage occurs in various locations (the mine pit, waste rock piles, etc) within the 15 km^2 area of the site. Consequently, a huge amount of water (~350m^3/h) containing radionuclides and heavy metals has to be treated continuously. There are many concerns among the local population about the potential environmental impacts of the site to both surface water and groundwater, as well as how the site may evolve over time [2, 3]. The aim of this research was to determine whether the radioactivity found in sediments along the Ribeirão das Antas River (~60 km) originated from natural or anthropogenic sources (especially from the uranium mine site). The sediment environmental compartment was chosen for this assessment since it is known to be a good historic record of contamination [4].

2. METHODS

2.1. Sampling strategy

Bottom sediments were collected in 2015 and 2016 at eight sampling stations located in reservoirs of the Antas river and at uranium minewater discharge points:

Station 1: Pitagueiras Creek, upstream of uranium mine;
Station 2: Águas Claras Reservoir, upstream of uranium mine;
Station 3: Discharge point of uranium minewater treatment plant;
Station 4: Águas Claras Reservoir, downstream of discharge point of uranium minewater treatment plant;
Station 5: Águas Claras Reservoir (near its outlet), downstream of uranium mine;
Station 6: Cipó Reservoir;
Station 7: Bortolan Reservoir;
Station 8: Rolador Reservoir.

2.2. Sampling procedure and sample treatment

A dredger drag, operated using a boat, was used for collecting bottom sediment in all the reservoirs. Samples were stored in plastic bags (20 kg maximum capacity) before being transported to the laboratory. Sample preparation, which involved sedimentation, siphoning, drying, milling and sieving, was carried out according to the standard operating procedure PN-LAPOC-8003 [5]. Samples were dried at 110°C until reaching constant weight, milled at 30 rpm in a stainless steel jar mill of 2.5 cm diameter and sieved at 1.7 mm (10 mesh).

2.3. High resolution gamma spectroscopy for determination of ^{226}Ra, ^{228}Ra, and ^{210}Pb

The activity concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb in the sediment samples were determined by high resolution gamma-ray spectroscopy following the standard operating procedure PN-LAPOC-8001 [6]. Dried samples were placed and sealed in metal boxes for 30 days — sufficient time to ensure secular radioactive equilibrium between ^{226}Ra and its progeny ^{214}Pb and ^{214}Bi. The gamma spectrometry analysis was carried out using a Canberra high purity germanium detector (relative efficiency 20%) and Genie 2000 spectral analysis software. For ^{226}Ra determination, the photopeaks used were those of ^{214}Bi (609 and 1020 keV) and ^{214}Pb (351 keV). The photopeak of ^{226}Ra (186.1 keV) was not considered owing to its low intensity and interference from the photopeak of ^{235}U (185.7 keV). The photopeak of ^{228}Ac with 6.12 h half-life (911 keV) was used for ^{228}Ra determination. For determination of ^{210}Pb, the photopeak 46.5 keV was used.

2.4. Inductively coupled plasma mass spectrometry (ICP-MS) for determination of uranium and thorium

For uranium and thorium determination, the sediment samples were first digested in nitric acid in a closed vessel using a microwave assisted digestion device following the procedure PN-LAPOC-7003 [7]. This standard operating procedure is similar to US Environmental Protection Agency Standard 3050B and provides partial samples, i.e. the non-aggregated bioavailable fraction of metals, especially those coming from anthropogenic contamination. After digestion, uranium and thorium were quantified by ICP-MS, according to standard operating procedure PN-LAPOC-7019 [8]. The technique involves sample ionization using an inductively coupled plasma, separation and quantification of the generated ions using a quadrupole mass spectrometer analyser. It is important to point out that the Chemical Analysis and Radon Laboratories at CNEN/LAPOC that carried out the analysis have participated in laboratory intercomparison exercises as well as in proficiency tests organized by national and international institutions and companies. In addition, these laboratories have implemented ISO/IEC 17025 for several of their assays and are currently in the process of accreditation by the Brazilian Accreditation Authority CGCRE/INMETRO.

3. RESULTS AND DISCUSSION

The results, expressed in terms of arithmetic mean and standard deviation, are summarized in Table 1. The values for ^{226}Ra, ^{228}Ra, ^{210}Pb and thorium at all sampling stations except sampling stations 3 and 4 can be considered as being typical of natural background levels for the region — activity concentrations in the same range as those presented here were found in similar studies in this area [4]. The values observed for uranium at sampling station 3 (settling sludge from the water treatment plant) are comparable with those obtained at sampling station 4 (bottom sediment in the Águas Claras reservoir). In fact, both materials present similar physical characteristics. The uranium concentration at sampling station 4 is quite unusual for natural bottom sediments and confirms an anthropogenic contribution. Moreover, this material is spreading over the Águas Caldas reservoir and concentrations of a few thousand becquerels per kilogram can be observed at its outlet located 2–3 km from sampling station 4 (downstream).

TABLE 1. SUMMARY OF RESULTS

Sampling station	Activity concentration (Bq/kg)				
	U	Th	Ra-226	Pb-210	Ra-228
1	140 ± 51	251 ± 59	147 ± 12	132 ± 28	240 ± 15
2	3070 ± 343	178 ± 30	240 ± 48	265 ± 44	241 ± 10
3	53139 ± 9806	373 ± 35	295 ± 29	<100	766 ± 20
4	50292 ± 11060	403 ± 70	506 ± 116	275 ± 54	567 ± 61
5	7446 ± 166	320 ± 58	324 ± 6	363 ± 32	376 ± 12
6	205 ± 92	268 ± 67	253 ± 100	358 ± 119	330 ± 13
7	451 ± 241	247 ± 35	255 ± 65	281 ± 61	268 ± 56
8	246 ± 18	235 ± 2	245 ± 120	327 ± 193	212 ± 6

4. CONCLUSIONS

Sediments at sampling stations 3, 4 and 5 (Águas Claras reservoir) exhibited significantly elevated concentrations of uranium, confirming an anthropogenic contribution. A strong dilution trend of this element is observed when comparing sampling stations 3 and 4 with the others downstream. A situation of environmental liability is observed and represented by these high concentrations, which come from the acid water treatment system and should be environmentally remediated. The high sulphate concentration in the acid water, which led to the precipitation of ^{226}Ra and ^{210}Pb, explains the low concentration of these two radionuclides in the sediment. The low thorium content in rocks located in the uranium mining site explains the low concentration level of this radionuclide and its progeny ^{228}Ra in the sediment.

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PRELIMINARY RADIOLOGICAL ASSESSMENT OF THE PHOSPHATE INDUSTRY OF SENEGAL

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Abstract

A preliminary assessment of radiation doses at workplaces in the phosphate industry was carried out in Senegal encompassing all phases, from the phosphate mine to the phosphoric acid production plant. Ambient radiation dose rates ranged from 0.04 μSv/h (the average background radiation dose rate) to 0.06 μSv/h in the open pit of the phosphate mine, up to a maximum of 12 μSv/h in the phosphoric acid plant. In the plant, radiation doses at some workplaces may give rise to an annual radiation exposure of 16 mSv. These radiation doses were mostly due to uranium series radionuclides with a smaller contribution also from radionuclides of the thorium series, both present in the phosphate rock. For example, the ^{226}Ra (U series) activity concentration in the phosphate rock was 1230 Bq/kg while the ^{228}Ra (Th series) concentration was 14 Bq/kg. Partitioning of radionuclides in phosphate materials during industrial processing was investigated and occupational radiation exposure was assessed.

1. INTRODUCTION

Sedimentary deposits of phosphate rock in the West African region are an important source of raw material for the world production of phosphoric acid and phosphate fertilizer. Several of these deposits occur in Senegal and most of the high grade deposits are concentrated in the west, where mining activities started in 1960 in Taïba. It is well known that phosphate rock contains, besides phosphorus and calcium, many other elements that are not desired in the composition of fertilizer. Such elements include uranium and its radioactive progeny which, together, may cause occupational exposure in phosphate processing facilities and enhancement of radioactivity in the environment [1, 2].

The most widespread industrial process for dissolving phosphate rock is attack with sulphuric acid, which is the basis for the production of phosphoric acid and leads to the production of significant quantities of phosphogypsum residues. There is a need for a deeper understanding of the radiological impacts associated with the various stages of phosphate processing and the various products. This preliminary assessment of radionuclide concentrations and radiation doses at workplaces was carried out in the phosphate mine and the phosphoric acid production plant at Taïba, in Senegal.

2. MATERIAL AND METHODS

The phosphate mining area of Taïba (15º07′46″N, 016º53′20″W), including the mine and the process plant, was visited. At several locations, external dose rates were measured, samples were collected and the GPS coordinates were duly recorded. Radiation measurements were performed at 1 m above the ground with a portable spectrometer Identifinder from FLIR, duly calibrated in a SSDL with standard ^{137}Ce and ^{60}Co sources. Representative samples of materials were collected with a large stainless steel spoon and sealed in identified plastic bags.

In the laboratory, the samples were dried in an oven at 60ºC and sieved through a metallic sieve in order to separate the materials with <63 µm grain size for the analysis. Aliquots of about 100g of these sample materials were analysed by gamma spectrometry in sealed boxes with the same geometry as that of customized multisource calibration sources from Eckert & Ziegler, using a BeGe large volume detector. Gamma spectra were analysed with Genie 2000 software and quality assurance was achieved through regular participation in the IAEA analytical inter-laboratory comparison programme and analysis of certified reference materials [3–5].

3. RESULTS AND DISCUSSION

Ambient radiation dose rates in the phosphate region, including the city of Taïba, ranged from 0.02 to 0.04 µSv/h. In the field, near the area of phosphate rock extraction, the ambient radiation dose rate above the untouched ground of sandy and non-mineralized soil was around 0.02 µSv/h. Phosphate extraction is performed by open pit mining, removing the upper soil layer to about 30 m depth in order to reach the phosphate rich layer. Phosphate is excavated from there and often the phosphate layer is immersed in the water table, which prevents dust release during excavation (Fig. 1). Radiation dose rates in the trench ranged from 0.39 to 0.65 µSv/h. From the mine, the phosphate ore is sent on a conveyor belt and by truck to the process plant at Taïba, where after grinding and sieving, a fine phosphate powder is allowed to react with sulphuric acid (hemi-hydrate method) to produce phosphoric acid.

FIG. 1. Phosphate rock mining (left); phosphoric acid plant and phosphogypsum piles at Taïba (right).

In the process plant, the ambient radiation dose rates near the ore piles were 0.62–0.64 µSv/h. Inside the facilities, dose rates increased to 6–8 µSv/h by the filtration unit where the slurry of rock and acid are filtered to separate phosphoric acid from the calcium sulphate dihydrate, known as phosphogypsum. The phosphogypsum is sent on a conveyor belt to waste piles where the dose rate reached 5 µSv/h. Phosphoric acid is stored and allowed to decant in stainless steel reservoirs and from there is pumped to the outside for loading into tank wagons and transported by rail to the phosphate fertilizer industry or to a harbour for export. Pipes and pumps used to transfer phosphoric acid develop scales inside, and need periodic cleaning for scale removal. External radiation dose rates at the surfaces of iron pipes reached 12 µSv/h.

Assuming an exposure time of 2000 h/a, the measured external dose rates would give rise to an annual dose of 2–4 mSv at several work stations, 16 mSv in the filtration unit and a maximum of 24 mSv at the surfaces of steel pipes. The results of sample analyses by gamma spectrometry are shown in Table 1. The activity concentrations of radionuclides in the ^{238}U, ^{235}U and ^{232}Th decay series were determined using the measured concentrations of ^{226}Ra, ^{235}U and ^{228}Ra, respectively. Although in the topsoil ^{226}Ra and ^{228}Ra were present in relatively similar concentrations (samples 1 and 2), in the phosphate ore the concentrations of uranium series radionuclides were much higher than those of thorium series radionuclides. The elevated uranium concentration and thus ^{226}Ra concentration is related to the marine origin of phosphate rock deposits. This phosphate rock was likely to have been produced by biogenic processes in an upwelling planktonic area, and its radioelement composition reflects the much higher abundance of uranium in sea water compared with that of thorium. Interestingly, the concentration of ^{40}K in the soil and the phosphate rock samples were all low — below the worldwide average for soils [4–8]. The predominant contribution to the enhanced external radiation doses in the process plant is therefore from the radionuclides of the uranium series.

TABLE 1. RADIONUCLIDE CONCENTRATIONS IN SOIL AND PHOSPHATE MATERIALS FROM THE REGION OF TAÏBA

	Activity concentration (Bq/kg dry weight)			
	^{40}K	^{226}Ra	^{228}Ra	^{235}U
Topsoil from the mining area	13 ± 6	16 ± 1	10 ± 1	<3.1
Taïba city, surface soil	18 ± 5	10 ± 1	8 ± 1	<3.1
Unprocessed phosphate rock	<22	1230 ± 50	14 ± 1	85 ± 10
Wet phosphate raw material	<19	1080 ± 80	8 ± 1	65 ± 12
Phosphogypsum	<19	600 ± 20	3 ± 1	14 ± 6

In the mine trench and in the facilities of the process plant, besides exposure to external radiation, the inhalation of dust containing radionuclides needs to be assessed as a pathway of radiation exposure, although inhalation of acid vapours might be a greater occupational health risk. Owing to the elevated concentrations of uranium series radionuclides, including ^{226}Ra, the radiological risk due to ^{222}Rn inhalation should be assessed.

Phosphogypum, the by-product of phosphoric acid production, is stockpiled in stacks near the process plant. The current content of ^{226}Ra in this material is 600 ± 20 Bq/kg, but other uranium series radionuclides, such as ^{210}Po and ^{210}Pb, are expected to be present in phosphogypsum in much higher concentrations because of their low solubility. The final destination of the phosphogypsum, and eventually its confinement, is open to assessment and decision.

4. CONCLUSIONS

This survey of phosphate rock mining and phosphoric acid production in Taïba identified enhanced radiation dose rates, especially in the process plant. A more detailed radiation protection study should be carried out in order to advise on radiation protection measures to be implemented by the company. In particular, the cleaning of pipes and disposal of scales, which may have a very high radium content, need specific attention.

Radon exposure in the facilities should be investigated also in order to identify any radiation protection measures for workers that might be necessary, besides protection against inhalation of acid fumes. In particular the large volume of phosphogypsum deposited in stacks in the surrounding environment, and its dispersal in the environment by rain and wind, needs assessment because of the close proximity to the city of Taïba. It is also pertinent to assess the radiological risk of the use of uranium rich phosphate fertilizers which may eventually advocate the relevance of extracting uranium from phosphoric acid.

Three of the main NORM industries identified worldwide, namely the oil and gas industry, the zircon industry and the phosphate fertilizer industry, are operating in Senegal. A more thorough radiation protection study should be carried out in order to increase regulatory awareness and advise on any radiation protection measures that should be implemented in these industries in Senegal. The management of NORM residues from these industries also needs due consideration and advanced planning.

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RADIOLOGICAL ASSESSMENT OF HEAVY-MINERAL SAND EXPLOITATION IN MOZAMBIQUE

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Abstract

Coastal deposits of heavy-mineral sand on the shore of the Indian Ocean are targeted by extraction industries intensively producing zircon, rutile, garnet, ilmenite and rare earth elements. Ambient radiation dose rates recorded in the areas of heavy-mineral sand deposits ranged from 0.4 µSv/h, the ambient radiation dose rate on undisturbed coastal dunes, to 3 µSv/h near the wet concentrate piles of heavy minerals prior to magnetic separation. In the facilities for magnetic separation of minerals, dose rates were around 1–3 µSv/h at workplaces and up to 8 µSv/h near the piles of segregated non-magnetic mineral fractions. Radionuclide analyses showed increasing concentrations of radionuclides from uranium and thorium decay series in segregated metal fractions, posing a risk of inhalation of dust particles with high radioactivity and exposure to segregated materials delivering high external radiation doses. Workers in the industrial separation and mineral storage facilities may receive doses exceeding the dose limit for members of the public. Therefore, radiation protection measures should be carefully implemented at workplaces and the radiological risk to members of the public should be assessed in the surrounding environment.

1. INTRODUCTION

Mozambique has extensive heavy-mineral sand deposits along its Indian Ocean coast and several mining projects are currently under development in that region [1]. The production of zircon and rare earth elements has been increasing in line with a growing demand from the microelectronics and telecommunications industries. These elements are often associated with radionuclides of natural origin in heavy minerals and this presence gives rise to a radiological risk that is important to quantify in order to facilitate the adoption of safety and health procedures to protect workers, members of the local population, non-human biota and the environment [2, 3]. A radiological investigation was performed at two areas, Moma and Angoche, in which heavy-mineral sand was being exploited (see Fig. 1).

2. MATERIALS AND METHODS

The two areas were visited and sampled jointly with inspectors of the Ministry of Mines of Mozambique in collaboration with safety officers of the mining companies. In both areas, radiation measurements were performed and samples collected for radiometric analysis in the laboratory. Measurement of ambient radiation doses was performed using a Thermo Scientific FH40 and a portable gamma spectrometer (FLIR) duly calibrated in a SSDL laboratory. Radiation dose surveys were carried out in the areas and triplicate radiation measurements were performed at specific spots, and recorded in conjunction with the GPS coordinates.

Samples of soil materials, sand from coastal dunes, and segregated heavy mineral fractions separated in the process plants were collected and placed in identified plastic bags. Later in the laboratory, these samples were dried in an oven at 60ºC to ensure standard drying, sieved through 63 µm Retsch sieves, and the fraction of grain size <63 µm retained for radioanalysis. Aliquots of these samples were analyzed by gamma spectrometry in the geometry of multisource customized standards supplied by Eickert & Ziegler. Other aliquots were used for determination of alpha emitting radionuclides by total dissolution with acids and microwave digestion, after addition of internal isotopic tracers according to analytical procedures described elsewhere [1, 4].

FIG. 1. Hydraulic mining of heavy minerals from coastal sand dunes at Angoche (left); arrival of heavy minerals wet concentrate at the magnetic separation plant at Moma coastal area (right).

3. RESULTS AND DISCUSSION

Ambient radiation doses measured over the sand dunes did not exceed 0.10 µSv/h and generally were below 0.8 µSv/h on the coastal dunes at Moma and Angoche. For comparison, the ambient radiation doses outside the coastal sand dune deposit, over clayey soils of the coastal plain in the Angoche area, were 0.04 µSv/h and lower.

At the mineral separation plants of both mining companies, in the area of the minerals feed, the ambient radiation doses increased to about 2.5 µSv/h and the highest radiation dose rates were measured near the piles of segregated minerals, reaching about 4.8 µSv/h near the non-magnetic minerals fraction pile and 2.05 µSv/h near the magnetic minerals fraction pile. The higest dose rate, 7.75 µSv/h, was measured near the non-magnetic piles after extraction of silica. Elsewhere in the facilities the external (ambient) radiation dose rates generally ranged between 0.10 and 3.10 µSv/h and radiation exposure at workplaces varied according to the location of work station and accumulation of mineral dust nearby.

Analysis of radionuclides in the fine fractions (<63 µm) of all samples confirmed that they contain radionuclides of the three naturally occurring radioactive series of ^{238}U, ^{235}U and ^{232}Th. The analysis results are shown in Table 1. Both in mass concentration and in activity concentration, the thorium series radionuclides were much more abundant than those from the uranium series. For example, in the unprocessed sand from dunes at Angoche, the thorium mass concentration was 0.25 ± 0.04 g/kg, while the total uranium (^{238}U alone accounts for 99.285% of the total mass of natural uranium) was 0.035 ± 0.001g/kg.

TABLE 1. CONCENTRATIONS OF RADIONUCLIDES AND ELEMENTS

	Site	Activity concentration (Bq/kg dry weight)		Mass concentration (g/kg dry weight)	
		^{238}U	^{226}Ra	Total U	Th
Sand dune: rich layer	Angoche #1	428 ± 16	324 ± 49	0.035 ± 0.001	0.25 ± 0.04
Rejected sand waste	Angoche #2	29 ± 2	24 ± 2	0.0024 ± 0.0001	0.0057± 0.0005
Sand dune: top layer	Angoche #5	40 ± 1	28 ± 2	0.0032 ± 0.0001	0.030 ± 0.002
Wet mineral concentrate	Angoche #6	727 ± 22	936 ± 110	0.059 ± 0.002	0.80 ± 0.08
Magnetic ilmenite	Angoche #7	86 ± 3	212 ± 17	0.0070 ± 0.0002	0.111 ±0.006
Non-magnetic fraction	Angoche #8	2729 ± 83	415 ± 42	0.221 ± 0.007	0.39 ± 0.02
Sand dune: top layer	Moma #1	9.3 ± 0.9	7.5 ± 0.6	$(7.6 \pm 0.8)\times10^{-4}$	0.0048±0.0003
Wet mineral concentrate	Moma #2	605 ± 22	353 ± 34	0.049 ± 0.002	0.34 ± 0.03
Magnetic fraction	Moma #5	179 ± 5	147 ± 13	0.0145 ± 0.0004	0.21 ± 0.02
Non-magnetic fraction	Moma #4	1560 ± 48	1076 ± 254	0.126 ± 0.004	1.1 ± 0.2
Standard zircon	Moma #11	1057 ± 30	800 ± 152	0.086 ± 0.002	0.054 ± 0.001

It was interesting to observe that uranium and thorium elements were present in the same proportions in most fractions separated in the industrial process, with the exception of the last product of the production chain, the ‘special zircon’. In this fraction, uranium became more concentrated than thorium (even more than in the standard zircon) due to tight association with zirconium in the heavier oxides. Most of the external radiation dose in excess of natural background that may cause exposure of workers will be due mainly to exposure to uranium and thorium series radionuclides present in the heavy minerals. In the facilities, the resuspension of ore particles creates a dusty atmosphere and workers may inhale dust containing radionuclides.

From the radiation dose measurements it is clear that the risk of external exposure of workers is high in some areas, especially where segregated materials are stored in large amounts. Air sampling and analysis of inhalable dust were not performed, but estimates can be made. In the dusty atmosphere of heavy minerals segregation, airborne dust levels may reach 10 mg/m^3. Applying the radionuclide concentration determined for ^{210}Po (2632 Bq/kg) the annual dose from inhaled dust may exceed 0.1 mSv. Inadvertent ingestion of dust may give rise to a similar dose from ingested radionuclides.

While in the area outside the facilities the ambient radiation dose would account for 350 µSv per year, in the average working conditions with an exposure of 1.5 µSv/h a factory worker may receive an annual dose of 3 mSv from external exposure plus 0.2 mSv from inhalation and ingestion. In the highest radiation dose rate area, by the mineral piles, the annual external radiation dose may reach 15.5 mSv for 2000 h exposure. These exposures would therefore significantly exceed the annual dose limit of 1 mSv above background for members of the public (including non-radiation workers). Therefore, in terms of international standards [5], the radiological risks associated with the operation of these industries are such as to require the classification of the facilities as radiation facilities and the application of radiation protection measures such as radiation monitoring, the hiring of a radiation protection officer, and inspections by radiation protection authorities. Due to the presence of dust in all areas of the factory, the use of respiratory protective gear should be enforced all times. Areas with higher radiation doses should be identified and monitored, and workplace hygiene should be managed to minimize radiation doses. In particular, frequent cleaning for removal of accumulated dust and minerals in the facilities will contribute to a reduction in exposure.

Several pathways such as water, suspended dust, agriculture products (particularly leafy vegetables) and coastal fish exist around the factories. These may transfer radionuclides from segregated heavy minerals to members of the public, but have not yet been assessed [6].

4. CONCLUSIONS

There is an association of thorium and uranium minerals with the heavy mineral fractions in coastal dune deposits in Mozambique, currently mined for zirconium and rare earth elements. Although in the unmodified coastal sand dunes the radiation background displays normal (average) radiation dose values, comparable to many other regions around the world, in the areas of industrial facilities with the segregation of heavy mineral fractions, uranium and thorium also become concentrated and radiation doses are enhanced. At some workplaces, the external radiation doses may reach and exceed internationally adopted annual radiation dose limits for members of the public. Occupational radiological risks associated with the inhalation of dust and ingestion of dust particles are present and do not appear to be negligible. Furthermore, dust dispersion in the atmosphere and disposal of radioactive ilmenite, a co-product of heavy mineral segregation with no commercial application in this situation, may create environmental and radiation impacts and may thus require proper management. Application of international radiation protection standards is needed in these industries.

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RESIDUES FROM NORM INDUSTRIES: A UNIFIED APPROACH TO ENVIRONMENTAL IMPACT ASSESSMENT

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Abstract

The graded approach to regulation entails not only an exposure assessment but also a demonstration that environmental criteria for long term human health protection are met. NORM activities cover a wide range of industrial sectors and involve various site specific and practice specific risk assessment methods. There are concerns that individually developed methods may lead to inconsistent results. This brings new challenges to industry operators and regulatory bodies and a generic assessment method based on well justified scientific guidance is needed. Considering possible exposure mechanisms, it is easy to identify those basic situations in which NORM may influence the environment seriously, namely: the existence of large amounts of solid process residue deposited into the environment and the release of contaminated water or air. Based on this, it can be concluded that all NORM situations can be covered by just three scenarios: disposal of solid residues on land, discharge of water into water bodies and stack emissions to the atmosphere. For each scenario, the crucial processes can be identified and a structured assessment process can be developed independently of any particular NORM industry. The proposed approach is illustrated in this paper using examples that include phosphogypsum stacks, the release of contaminated water from coal mines and stack emissions from coal fired power plants.

1. INTRODUCTION

In managing the process residues from NORM industries, protection of the public and the environment needs to be addressed. The graded approach to regulation entails not only the assessment of short term human exposure but also a demonstration that environmental criteria for long term human health protection are met. Notwithstanding the clearance levels established by the IAEA [1] and implemented into European legislation [2], situations in which the presence of radionuclides of natural origin in water sources may affect the quality of drinking water supplies or any other pathways of exposure may require notification to the regulatory body.

Since NORM activities cover a wide range of industrial sectors with diverse characteristics, various site specific and practice specific risk assessment methods are used. Bearing in mind that many of the industries in question have not been regulated in terms of radiation protection until now, concerns exist that individually developed methods may lead to inconsistent results. This brings new challenges to industry operators and regulatory bodies and a generic assessment method based on well justified scientific guidance is needed.

NORM is usually encountered in the form of residues or by-products from minerals processing activities, which are often classified into a special category of radioactive material. Even though the presence of NORM is quite widespread, the incremental (above background) radiological risks to individuals are largely limited to risks to groups of workers involved in the various processing activities, and thus mostly within the premises of the plant. The impact of a NORM residue beyond the premises of the plant depends very much on the type of residue, in particular whether it is in the form of a solid, liquid or gas:

(1) For a NORM residue in solid form, the impact on the surrounding ecosystem, even close to the facility, tends to be limited in reality, especially when one considers that most NORM facilities are located in industrialized environments that are already significantly altered by human activity. A slightly different situation exists in the case of so-called legacy sites, where NORM may have been disposed of at a time when there was little or no appreciation of natural radioactivity or environmental issues. Irrespective of any historical factors, however, it goes without saying that solid NORM residues are usually very abundant compared with 'classical' radioactive waste such as that from spent radioactive sources or even spent nuclear fuel, and has completely different characteristics. On the other hand, it is usually accumulated in relatively small, well-defined areas and the extent of contamination is very limited in comparison with areas that can be contaminated by radioactive fallout, for instance after a nuclear accident.

(2) A NORM residue in liquid or gaseous form is usually discharged to the environment as a process waste. The environmental impact of such discharges may be more widespread and serious than that associated with solid NORM residues, even at distances far from a discharge point, but it is of course far less visible.

From the above mentioned considerations, it is clear that not all NORM related scenarios have serious implications for the environment. Moreover, it is quite easy to identify those basic scenarios for which NORM may indeed give rise to significant environmental concerns — these are following:

(i) The existence of very large amounts of solid waste deposited directly into the environment,
(ii) The release of contaminated water into water bodies,
(iii) The release of contaminated air or airborne dust into the atmosphere.

After applying these scenarios to the list of NORM industries that have been identified either on the basis of possible risk to workers or according to the waste catalogue, the list of situations of concern shrinks markedly and the criteria needed for environmental impact assessment can be easily defined.

2. SOLID NORM RESIDUES

2.1. Main NORM industries to be considered

The presence of enormous amounts of solid NORM residues often leads to them being regarded as the greatest contributor to the environmental burden related to NORM. But actually there are not so many industries that finally produce solid NORM residues in such quantities. The first and the most common are coal fired power plants that produce very large amounts of coal combustion products, mostly in the form of flyash. In the European Union, where in fact a significant part of electricity generation still comes from coal [3], the total amount of coal combustion products being generated annually is more than 100 million t [4]. But the radiological problem from an environmental contamination point of view is not so great when one considers that the radionuclide activity concentrations are usually less than 200 Bq/kg. Moreover, coal combustion products, especially flyash, are being increasingly regarded as valuable raw materials for many purposes, so that their disposal on land is slowly diminishing. In fact, the radioactivity content is so low that the only limiting factor for the use of coal combustion products as by-products is that, for the construction of buildings, they should not be used directly (i.e. without mixing with other, less radioactive material).

The second major producer of solid NORM residues is the phosphate industry, which in many countries around the world is engaged in the production of fertilizer and other products from phosphate rock, mainly via the intermediate product phosphoric acid. The phosphoric acid production process generates about 4 t of phosphogypsum (PG) for every tonne of phosphoric acid produced. This residue is mostly deposited on the land as 'stacks' A typical PG stack covers several hectares of land and contains millions of tonnes of residue. Although some by-product uses of PG have been identified, the amounts of PG already in existence and still being generated are so large that it is difficult to see how such by-product uses can, in the short term, bring about any significant reduction in the amounts of PG deposited as waste. In comparison with coal ash, the radionuclides are present in higher activity concentrations and are not in equilibrium. However, the radioactivity content is still moderate and any adverse effects of PG on the environment are due to its chemical composition rather than to its radiological properties.

Other solid NORM residues that may be abundant enough to have a significant impact on the environment include those from the following NORM industries:

(a) The production of non-ferrous metals such as tin and niobium.
(b) The production of rare earth elements from heavy mineral sands found on or near beaches. Most of the radioactivity is contained in the monazite fraction, which is separated from the heavy mineral concentrate and further processed to extract the rare earth elements. After separation of the various mineral fractions, (including the monazite fraction), the residue usually contains less radioactivity than the original sand excavated. Since this residue is regarded as a waste, however, its return to the original beach of origin may be forbidden, depending on legal restrictions in the country concerned.
(c) The mining and processing of uranium ore is a source of solid NORM in the form of gangue or of excavated uranium ore that is not of a high enough grade to be processed, as well as radium-containing tailings from the uranium extraction process.

2.2. Assessment criteria

From an environmental point of view all the above mentioned examples of solid NORM residues are very similar. The area inside the perimeter of a NORM residue deposit is completely changed and usually radioactivity is not a major pollutant considering the high amount of alien material deposited. Even if an ecosystem had been present initially, it would have disappeared after the residue deposit was created.

Depending on the material deposited, a secondary succession can occur over time and a kind of simple ecosystem can appear again, especially if supported by human activity. In such a case, a residue heap covered with vegetation can create a good test field for observation of external exposure, transfer factors to biota, accumulation in biota and finally the effect on biota. However, it does not represent a real ecosystem and observations cannot be easily generalized. This is especially so when taking into consideration that other biological endpoints caused by a stress originating from the seriously altered habitat can play a greater role. Moreover, even if one considers the largest existing NORM residue deposit, the observed ecosystem, if revitalized, will inevitably be limited to plants, invertebrates, amphibians or mammals that occupy a small habitat that does not extend beyond the perimeter of the deposit.

In terms of environmental protection, the only thing that can be analysed is the different ways in which the residue or radionuclides within it are released from a deposit and dispersed into a surrounding environment that could actually be regarded as an ecosystem. Again, however, such residue deposits do not usually represent an area that is unaffected by previous human activity. It is certain that, for instance, the transport (at least) of such large amounts of bulk material will change the ecosystem seriously, even more than the effect of an elevated level of radioactivity, and the situation will continue to have the characteristics of a heap area. Apart from this, when analysing such a case, processes leading finally to the migration of radionuclides from the deposit to nearby or distant areas are the most important.

The first type of process responsible for NORM dispersion consists of different kinds of erosion when NORM particulates are transported from a deposit into the surrounding environment. In this case, the physical NORM properties are the most important. These processes can eventually lead to contamination of the surrounding land area and (rarely and temporarily) the quality of the atmosphere.

The second type of process is leaching, when the residue is exposed to environmental conditions. In this case, the chemical properties of the residue, and particularly the radionuclide species, are important. Unlike in the case of erosion, leaching of NORM must be considered not only on the surface but also below a deposit, meaning that groundwater contamination should be taken into account. However, this aspect is ultimately more important in terms of risk to humans rather than to the environment. Having defined the main processes, all the derived parameters necessary for risk assessment can be easily identified.

3. LIQUID NORM

3.1. Main NORM industries to be considered

In the case of water release, the list of NORM industries is rather short and first place on this list is occupied by crude oil and natural gas exploitation, including recently developed technology to make the shale gas resource available [5–8]. Huge amounts of radium rich water (referred to as process water or produced water) is released into the environment, especially the marine environment in the case of offshore platforms [9].

The second industry on the list is the underground coal mining industry where formation water with a high radium content is also often present [10]. However, in comparison with the oil industry this problem is not well known. Well documented data are available only from the Polish mining industry where about 40% of still active coal mines discharge water with radium activity concentrations at levels up to a few hundred becquerels per litre. There are no well justified reasons why such a situation does not exist also in other countries that excavate much more coal than Poland (coal is always excavated from carboniferous strata, no matter where in the world it is). Besides fossil fuel exploitation, radium rich water often occurs when geothermal water is used either for energy generation or in a spa, but usually such waters are not as abundant as in the mining industry and the radiation risk to the environment is rather limited [11].

A different case of NORM contaminated water concerns the water released from uranium mining that contains a significant amount of uranium. There are many examples of uncontrolled release that took place in the past, when no one was considering environmental protection. Similar situations are also observed at a few sites in the world where radium had once been produced.

3.2. Assessment criteria

In the case of liquid NORM, the probability of an impact on the environment is greater when compared with solid NORM and the exposure scenario is more complex. In general, three considerably different exposure scenarios need to be analysed: releases into a marine ecosystem, releases into inland running water and releases into stagnant water. The most important processes influencing the behaviour of NORM released in water are dilution and precipitation, irrespective of the particular release scenario.

3.2.1. Discharges to the sea

In this scenario, contaminated water is discharged directly to the sea (e.g. from an offshore oil platform). The first obviously identified process is dilution of radioactivity in the sea water. It is quite convenient to assess the whole problem based on this process. When one takes into account the amounts of discharged process water and of surrounding sea water, the expected effect (viewed simply as sea water contamination) assessed in this way is eventually negligible.

However, this is a very superficial approach and in order to evaluate possible effects on the marine ecosystem some secondary processes that can lead to radionuclide accumulation should be analysed. Available data have shown that, particularly in the case of radium rich discharge water, precipitation processes can result in sediments with a high content of radionuclides. This process is very efficient and depends on changes of pressure and temperature of water released from a rig and its chemical composition. Sediments with enhanced radium concentration then start a series of processes that usually are considered in any radiation risk scenario, i.e. external exposure, migration, resuspension, etc.

3.2.2. Discharges to watercourses

As in the case of discharge to the sea, the main concern is associated with the discharge of radium rich water In the coal mining industry, where the extraction process is completely different from that for oil and gas and all formation water flowing into the underground galleries must be pumped to the surface, the main process influencing the environmental behaviour of radium in the mine water is radium precipitation. Depending on the particular situation, radium can start to spontaneously precipitate next to the inflow point in underground galleries and immediately lead to the creation of sediments. As the efficiency of the precipitation process depends on the total radium, barium and sulphate balance and not all of the radium is removed immediately, some radium rich waters are discharged more or less directly into surface fresh water.

Dilution is then again identified as a primary process that should be considered in conjunction with secondary processes similar to those for discharges into the marine environment, i.e. accumulation either in bottom sediments or in biota. After mixing with inland water, precipitation or co-precipitation becomes less likely, owing to the fact that an excess of radium solubility product (even when barium ions are present) is almost impossible. Different sorption processes must be considered in order to predict radionuclide accumulation in bottom sediments.

It is worth mentioning that in all the reported cases of radium rich water discharge, radium isotopes are present in ionic form. No radioactive particulates are present in such waters. That is why the process related to translocation of suspended matter with formation water does not influence contamination propagation. Later, after sediments have been created, processes related to water erosion also should be considered. In watercourses with fast flowing water, contaminated sediments usually are diluted and fractionation leading to additional accumulation is not observed.

Actually, when considering contamination translocation with solids, a human activity should be considered first — as many examples have shown, contaminated bottom sediments have often been removed in order to keep the mine dewatering system working properly, especially in the case of smaller water courses. In the past, when no one was warned about radioactive contamination, sediments were relocated into neighbouring arable land, forest or other local ecosystem. This resulted in some hot spots where relatively high contamination is now observed. Similar effects also resulted from mine dewatering system failures when some small areas became flooded by mine water. In both cases the possibility to observe a more or less natural terrestrial ecosystem contaminated by radium exists. But actually all the contaminated sites referred to above are so small that possible effects on biota can be observed only up to the level of small rodents, whose habitats do not exceed the perimeter of the site concerned. Finally no one should expect serious biological effects on larger populations occurring across the whole area where contaminated spots exist.

3.2.3. Discharges to (settling) ponds

A special case can be observed when the process of precipitation and then sedimentation lead to long term accumulation of radionuclides. Unlike watercourses where, for various reasons, contaminated sediment can be moved, a pond when fed with contaminated water can become an efficient pollutant 'repository'. It is a well known phenomenon and actually bottom sediments from a lake are frequently used for the purpose of the current state assessment of an environment not only when radioactivity is investigated. Conducive circumstances exist in the mining industry where settling ponds are used for the preliminary purification of mine water from suspended matter before its release into a watercourse. A side effect observed is radium accumulation in bottom sediments. Currently such reservoirs are artificial ones constructed specially for such purpose but in the past natural ponds were often adapted for this purpose.

Artificial settling ponds usually are of limited capacity and often must be cleared from accumulated sediments. Final disposal of such waste is a separate case, but from an environmental point of view it can eventually be treated as a case of a waste deposit as discussed above.

Natural lakes usually are large enough to be exploited for a much longer time and final cleaning processes (i.e. removal of contaminated bottom sediments) are not justified from technological and economic points of view. When such a lake is still used for mine water retention, the exposure scenario is similar to a waste deposit i.e. a core that is heavily contaminated (not only with radioactivity but also with heavy metals and, most importantly, with a high content of salt) and possible effects on the more or less natural surroundings. Of course, migration processes slightly different from those for land disposal must be taken into account. However, when mine water discharge is stopped the situation can change rapidly. Owing to natural processes (slowly) or intentional technical activity (quickly), water can be cleaned and secondary biological succession can occur immediately. Finally a lake ecosystem can be regenerated under continuous exposure to radionuclides accumulated in bottom sediments. However, such a situation can be considered as an environmental risk — from a logical point of view the situation is very similar to that existing at a solid NORM waste deposit discussed above. The affected ecosystem is limited and already anthropologically changed. Such a kind of exposure scenario is rare and from a regulatory point of view is considered together with discharges into watercourses that are influencing the environment more.

4. STACK EMISSIONS OF NORM

4.1. Main NORM industries to be considered

Among all the identified NORM activities, this kind of emission and the related exposure scenario is the most frequently observed. It is a consequence of high temperature processes for energy generation and processing of metal ores. Every coal fired power plant emits into the atmosphere some amount of radionuclides of natural origin, even if effective systems for exhaust gas cleaning have been applied. But in comparison with other NORM sources the list of radionuclides that should be considered in such a scenario is limited to elements characterized by low boiling temperatures. In practice, the only radionuclide in such cases is ^{210}Pb. In principle, ^{210}Po should also be considered but due to difficulties in the measurement (it is a pure alpha emitter) not so many data are available on its presence in the air just next to a stack. Other sources of such emission are ore sintering and metal smelting plants where blast or arc furnaces are applied. Currently all such processes are well equipped with filtration systems and, from a radiation protection point of view, the solid waste (dust) finally collected is more important than direct emission to air. An additional difficulty that effectively masks the possible effects caused by stack emission is the natural presence of ^{210}Pb in the atmosphere caused by radon exhalation from the Earth's surface. Under natural conditions, the activity concentrations of ^{210}Pb can reach levels of millibecquerels per cubic metre, but next to a stack and depending on the wind an excess of lead can be observed. This can be demonstrated by measurement of the lead to polonium activity ratio.

4.2. Assessment criteria

Irrespective of the source of ^{210}Pb in air, the main process of importance when possible environmental effects are considered is the deposition of this radionuclide on the Earth's surface. In fact, in the thin top layer of undisturbed soil, the ^{210}Pb activity concentration is slightly higher than that expected from the decay of the parent radionuclide ^{226}Ra (in spite of the fact that some radon will have been exhaled). The average retention time of ^{210}Pb created by radon decay in air is about 6 d. Apparently, the deposition process is influenced by meteorological conditions and common models used for radioactive fallout can be applied in order to observe the possible contribution of ^{210}Pb emitted from industry to the total amount of this radionuclide present in the top layer of soil. However, the total final activity concentration increment observed is negligible when considered as source of additional exposure.

The second process related to the presence of ^{210}Pb in air is interception of this radionuclide by plants. About 90% of this radionuclide present in above ground plant tissue has been incorporated in this way. Again, the contribution of 'additional' lead originating from industrial processes is not so important when compared with the naturally observed levels. In general the total effect of the release of lead and polonium from industrial activity is masked by the natural migration of these nuclides in the environment. However, it must be underlined that final accumulation of these radionuclides in biota is not negligible from a radiation protection point of view. On the other hand, the problem with NORM in such branches of industry is rather related to the disposal or reprocessing of dust collected on filters.

5. CONCLUSION

Considering possible interactions, the environmental burden imposed by NORM industries can actually be restricted to a few exposure scenarios and it is quite easy to identify the basic situations when NORM influences the environment seriously, namely, the existence of enormous amounts of waste deposited directly into the environment, the release of contaminated water and the release of contaminated air or dust. It can therefore be concluded that three exposure scenarios cover all NORM situations: disposal of NORM residues on land, discharge of contaminated water into water bodies and stack emissions of contaminated air and dust into the atmosphere. Then, having such a categorization, crucial processes governing radionuclide behaviour can easily be identified for each scenario and a structured assessment process can be developed independently of any particular NORM industry. Applying this approach, the notification process in accordance with new regulatory requirements can be formalized and transparent criteria can be applied for all stakeholders involved.

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Poster Prsentation

NATURAL URANIUM IN BIOLOGICAL MATERIAL FROM HIGH BACKGROUND RADIATION AREAS IN BRAZIL

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Abstract

Through the analysis of 319 samples, the activity concentrations of natural uranium were determined in 15 different biological materials from high background radiation areas of Brazil in which are situated four industrial facilities associated with NORM in different ecosystems. The uranium concentrations ranged from 0.0474 Bq/g in milk to 0.3278 Bq/g in cara fish. Three groupings of activity concentration were identified using statistical methods.

1. INTRODUCTION

Brazil has the world’s sixth largest uranium reserves [1] and several areas of the country can be characterized as high background radiation areas. Four industrial facilities associated with NORM, owned by Brazilian Nuclear Industries and located in different ecosystems, were investigated:

(1) The Uranium Concentrate Unit (URA), a uranium mine in operation, situated in a savanna–caatinga ecosystem;
(2) The Unit of Santa Quitéria (USQ), a phosphate deposit with associated uranium and still in the commissioning phase, situated in a caatinga–cerrado ecosystem;
(3) The Heavy Metals Unit (UMP) operating in an Atlantic rain forest ecosystem associated with coastal lagoons;
(4) The Ore Treatment Unit (UTM), a closed uranium mine in the decommissioning phase, located in a rain forest and cerrado ecosystem.

An industrial facility involving NORM is a planned exposure situation, that is, a situation of exposure involving the deliberate introduction and operation of a radioactive source [2]. As such, mining and minerals processing activities associated with NORM are regulated in Brazil by the regulatory standard CNEN-NN-4:01 [3] and others [4–7]. Such activities require an environmental radiological monitoring programme (abbreviated to PMRA in Portuguese) with specific matrices and frequencies of sampling [7]. The study of radionuclide behaviour in the environment, a part of radioecology [8], is an important tool that allows a PMRA to be established. The PMRA is performed as a tool to assess the environmental impact of the radiological practice. To assess the radiological environmental impact of a practice, parameters other than just activity concentration are important. Among such parameters, the most important ones are the use of biological media by the population, assessed by their consumption rate, and the dose conversion factors of the radionuclides concerned. In order to contribute to environmental radiation protection and to radioecological knowledge of uranium in biological material from areas associated with NORM industrial activity, this study aims to assess the activity concentrations of natural uranium in 15 different biological materials from several environmental compartments in four areas of industrial activity involving NORM.

2. MATERIALS AND METHODS

The industrial facilities are situated at the following locations:

(i) URA is in the rural area of Caetité, southwest of Bahiathe;
(ii) UTM is in the city of Caldas, Minas Gerais;
(iii) USQ is in the municipality of Santa Quitéria, Ceará;
(iv) UMP is in the city of Campos dos Goytacazes, Rio de Janeiro.

Samples of the biological material were collected in the vicinity of the facilities, dried to a constant weight at a temperature below 90°C and calcined until light coloured ash was obtained. An aliquot of the material was treated at a temperature below 80°C with a mixture of concentrated nitric acid and hydrogen peroxide until the material had become completely dissolved The U_{nat} activity concentration in the dissolved material (in terms of wet weight) was measured at the environmental laboratory of UTM using the method described in Ref. [9], which involves extraction by organic solvent followed by spectrophotometry.

A statistical analysis of the results was performed, generating data such as mean, coefficient of variation and number of samples [10–11]. An analysis of variance (ANOVA) was performed [12–14] to compare the mean activity concentrations of each biological material. The rejection value of P(α) was 0.05. When differences between the mean activity concentrations were observed, a Tukey test was performed to divide the mean values into groups [12–14].

3. RESULTS AND DISCUSSION

The results of the statistical analysis of the U_{nat} activity concentrations are shown in Table 1 and Fig. 1. The mean activity concentrations ranged from 0.0474 Bq/g in milk to 0.3278 Bq/g in cara fish, corresponding to a mean activity concentration range of 0.2804 Bq/g. The coefficients of variation in the biological materials ranged from 31 (potato) to 160 (vegetation). The high variability of the vegetation matrix can be partly explained by its composition, since such vegetation consists of leaves of different tree species found in the region, while the low variability for potatoes may be partly explained by the fact that only five samples were involved. The ANOVA exercise resulted in a P value tending to zero, associated with an F value of 6.21. Thus, statistically significant differences existed between the mean activity concentrations observed for the various biological materials. As shown in Table 1, the application of the Tukey test divided the biological materials into groups A, B or C depending on whether they were identified as having high, medium or low activity concentrations, respectively. Cara fish fell within group A (high mean activity concentration), while pasture, palm, corn, sugar cane, beans and milk all fell within group C (low mean activity concentration).

TABLE 1. NATURAL URANIUM CONTENT IN BIOLOGICAL SAMPLES

	No. of samples	Mean U_{nat} activity concentration				
		Value (Bq/g)	Coefficient of variation (%)	Tukey test grouping		
Cara fish	9	0.3278	45	A		
Tilapia fish	14	0.1860	86		B	
Lambari fish	8	0.1100	81		B	C
Silage	11	0.1050	42		B	C
Manioc flour	16	0.0991	78		B	C
Potato	5	0.0946	31		B	C
Manioc	45	0.0943	137		B	C
Vegetation	23	0.0901	160		B	C
Traira fish	5	0.0802	105		B	C
Pasture	39	0.0767	81			C
Palm	11	0.0721	67			C
Corn	27	0.0702	113			C
Sugar cane	12	0.0685	81			C
Bean	21	0.0557	88			C
Milk	73	0.0474	107			C

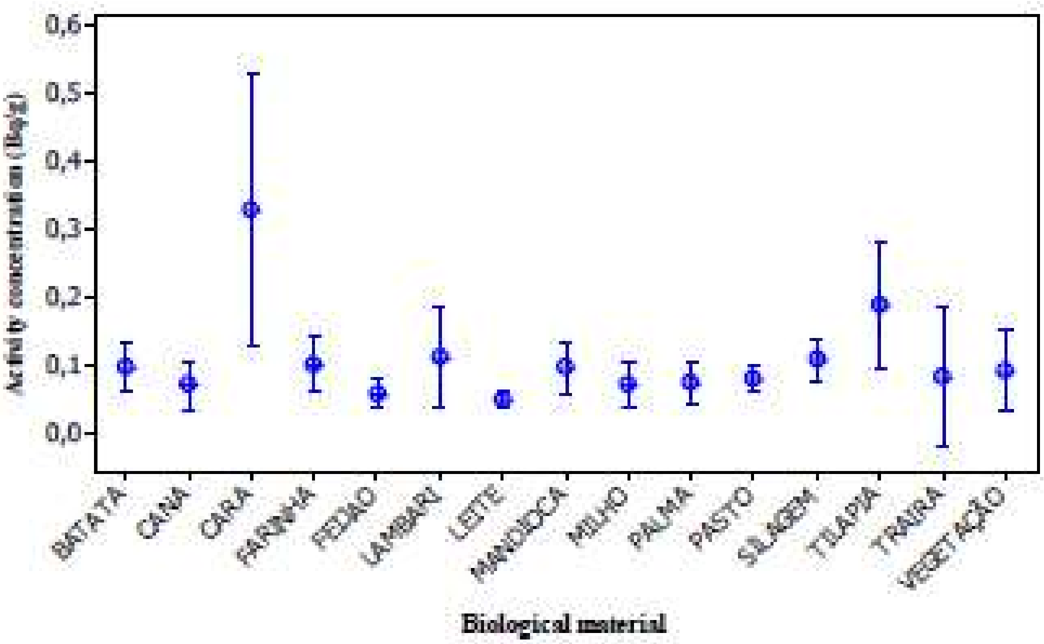

FIG. 1. Mean and confidence interval of activity concentration of natural uranium.

4. CONCLUSIONS

From a radioecology point of view, it can be concluded that:

(a) The biological materials can be divided into three groups according to their U_{nat} content (high, medium or low activity concentration);
(b) The various biological species have different accumulation efficiencies;
(c) There is a tendency for fish to accumulate uranium more efficiently than the other types of biological species;
(d) The grinding and dewatering of manioc to produce manioc flour does not have any significant affect on the uranium content.

From an environmental radiation protection point of view, the picture is more complicated than simply the existence of three groups of uranium activity concentration. The rate of consumption by members of the public also needs to be taken into account, since this is a key determinant of exposure and therefore radiological risk. Taking this into account, the assessment of biological materials according to the associated risk needs to be investigated and other radionuclides need to be included in this assessment.

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Poster Presentation

NORM WASTE FROM GHANAIAN OIL PRODUCTION

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Abstract

The management of NORM waste such as produced water and scale arising from oil and gas exploration and production in coastal areas is recognized universally as an important issue. Knowledge of the radiological characteristics of such waste is needed in order to assess any associated health risks. However, data on NORM waste from oil and gas production, particularly in Africa, are incomplete. In some areas of Africa (e.g. the Gulf of Guinea) such information is almost totally missing and although monitoring programmes are being undertaken very little is known about contaminant levels and trends. This paper describes an assessment of the activity concentrations and distribution of several radionuclides in the ^{238}U and ^{232}Th decay series, and ^{40}K, and the physico-chemical characteristics of produced water and scale from Ghanaian offshore production oilfield platforms. The use of a combination of the analytical techniques of non-destructive gamma spectrometry and alpha spectrometry after radiochemical separation (as well as scanning electron microscopy) has resulted in a more precise radiochemical and radiological characterization of produced water and scales.

1. INTRODUCTION

Until now, no radiological surveys of the oil and gas production facilities and surrounding areas at the Jubilee oilfields at the West Cape Three Points in Ghana, as well as the Saltpond offshore field, have been carried out. Consequently, radiological data on the NORM waste generated are unavailable This situation could be of great concern in terms of radiation protection of workers, members of the public and the environment, particularly as Ghana has been producing oil in commercial quantities for the past few years. The work described in this paper is a first step in the assessment of the radiological hazards and risks to members of the public and workers due to the oil production and associated waste management activities.

2. MATERIALS

NORM waste in the form of scale and produced water arising from crude oil production activities from the Saltpond and Jubilee oilfields of Ghana have been radiologically assessed by measurements of the activity concentrations of ^{234}U, ^{238}U, ^{210}Po, ^{230}Th, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{234}Th, ^{228}Ra, ^{228}Th, ^{224}Ra and ^{40}K. The two waste types that have been analysed can be characterized as follows:

(1) Scale — generally in the form of barium sulphate, barium carbonate, calcium carbonate and associated radium compounds — originates from the reservoir underground formations and bedrock and is carried by the oil and production fluids to the surface. Changes in pressure, temperature and/or other parameters cause the scale to precipitate and adhere to the internal surfaces of the pipes. The presence of radium and its decay products in the scale (which depends on factors such as the type of geological formation and the age of the well) introduces the possibility of radiological risks for workers, members of the public and the environment during the removal, storage and disposal of the scale.

(2) Produced water represents the largest volume of waste associated with oil and gas production. It consists of formation water, injected water (both of which are carried to the surface with the oil), small volumes of condensed water and any chemicals added downhole or during the oil/water separation process. Despite treatment prior to discharge to satisfy regulatory restrictions, which are mainly concerned with the oil content, produced water contains radionuclides such as ^{226}Ra and ^{228}Ra. While these radionuclides are present at low concentrations, they are difficult to remove and usually end up being discharged with the water. This gives rise to possible risks to humans and the environment.

2. METHOD

The activity concentration measurements were performed using the techniques of (i) non-destructive gamma spectrometry and (ii) alpha spectrometry after radiochemical separation. Scanning electron microscopy and other complementary techniques have also been used.

All samples were non-destructively analysed by high resolution gamma spectrometry using a p-type extended range germanium coaxial detector (XtRa) with a relative efficiency of 37.1% and with an energy resolution of 1.8 keV for the ^{60}Co gamma-ray energy peak of 1332 keV. The detector was housed within 10 cm passive shielding of ancient lead, while active shielding made with an organic scintillation detector (Bicron BC-418) was placed on the top of the lead shield, working in anti-coincident mode with the germanium detector. This allowed remarkable precision in the environmental gamma radiation measurements due to the very low background. The identification of individual radionuclides was performed using their characteristic gamma-ray energies and the quantitative analysis of radionuclides was performed using the Genie 2000 gamma acquisition and analysis software. Background spectra were acquired and used to correct the net peak area of gamma rays of the measured isotopes. A multi-gamma certified cocktail standard (^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y), spiked either onto water (for produced water) or gypsum (for scale) prepared in the measuring geometries was used for efficiency calibration of the gamma system. In calculating the activities, considerations of coincidence summing factors were incorporated.

The alpha emitting radionuclides ^{238}U, ^{234}U, ^{230}Th and ^{210}Po were determined in representative samples of produced water and scales by alpha-particle spectrometry. The sample preparation was carried out in three main steps: (i) digestion (solid) or preconcentration (liquid), (ii) radiochemical separation and (iii) electrodeposition. The details of these procedures can be found in Ref. [1]. Source preparation for ^{238}U, ^{234}U, ^{230}Th analysis was done by independent electrodeposition onto stainless steel discs, applying the method described in Ref. [2], while for ^{210}Po analysis it was done by self-deposition onto copper discs. The alpha measurements were made using passivated implanted planar silicon (PIPS) detectors (active surface 450 mm^2) installed in a Canberra 8-chamber Alpha Analyst system. Each chamber was devoted to the exclusive measurement of one element in order to avoid cross contamination. The measurements were carried out at a source to detector distance of 0.5 cm. The accumulation and analysis of alpha spectra were done using Genie 2000 software with a measurement time of 200 000 s. The background spectrum was also used to determine the minimum detectable activity (95% confidence level) of the U, Th and Po isotopes (~0.1 mBq) for a measuring time of 2–3 d. The mean recovery yields of the radiochemical procedure for the analysed samples were 68% for U, 80% for Th and 42% for Po.

3. RESULTS

3.1. Produced water

The activity concentrations of ^{226}Ra, ^{228}Ra, ^{228}Th, ^{224}Ra and ^{40}K in produced water samples from the two oilfields, determined by gamma-ray spectrometry, are presented in Table 1. The produced water samples had been collected from oil/water separators prior to discharge to marine waters from both the Saltpond and Jubilee oilfields platforms. The average value of the activity concentration of ^{226}Ra was 15.2 ± 7.1 Bq/L (range 6.2–22.2 Bq/L), while for ^{228}Ra the average activity concentration was 23.0 ± 13.5 Bq/L (range 6.4–34.2 Bq/L). A comparison of the mean activity concentrations of radionuclides for the two oilfields indicated that the activity concentrations for produced water from the Saltpond field were three times higher than those for the Jubilee field. This can be attributed to factors such as the maturity of the Saltpond field compared with the Jubilee field, the geological characteristics of the reservoir rocks, the type of hydrocarbons produced and the operating conditions for the oilfields. For both oilfields, however, the average concentrations of ^{226}Ra and ^{228}Ra for were well within the reported worldwide range of 0.002–1200 Bq/L for ^{226}Ra and 0.3–180 Bq/L for ^{228}Ra in produced water [3].

TABLE 1. GAMMA EMITTING RADIONUCLIDES IN PRODUCED WATER

Sample code	Activity concentration Bq/L				
	Ra-226	Ra-228	Th-228	Ra-224	K-40
Jubilee field:					
JF1	6.7 ± 0.2	6.6 ± 0.2	0.82 ± 0.01	0.82 ± 0.01	6.3 ± 0.5
JF2	7.6 ± 0.3	6.9 ± 0.1	1.22 ± 0.03	1.43 ± 0.04	8.3 ± 0.8
JF3	6.2 ± 0.1	6.4 ± 0.1	0.71 ± 0.01	0.69 ± 0.01	7.7 ± 0.7
JF4	6.6 ± 0.1	6.6 ± 0.1	0.81 ± 0.01	0.78 ± 0.01	5.9 ± 0.4
JF5	6.8 ± 0.2	6.7 ± 0.1	0.92 ± 0.01	0.92 ± 0.02	7.3 ± 0.5
Saltpond field:					
SF6	20.1 ± 0.5	33.5 ± 1.0	5.7 ± 0.2	5.8 ± 0.3	23.4 ± 2.4
SF7	22.2 ± 0.9	34.2 ± 1.6	5.5 ± 0.2	5.4 ± 0.1	22.3 ± 2.3
SF8	19.5 ± 0.5	32.3 ± 0.9	5.0 ± 0.2	5.1 ± 0.1	22.1 ± 2.3
SF9	22.1 ± 0.5	33.6 ± 1.5	5.4 ± 0.2	5.6 ± 0.2	22.2 ± 2.4
SF10	19.7 ± 0.8	33.1 ± 1.0	6.0 ± 0.2	6.0 ± 0.3	22.3 ± 2.5

The activity concentrations of ^{234}U, ^{238}U, ^{210}Po, ^{230}Th and ^{232}Th were determined in the same produced water samples by alpha spectrometry. The ranges for each radionuclide were as follows:

^{234}U: Below the lower limit of detection up to 0.0061 Bq/L,
^{238}U: Below the lower limit of detection up to 0.0055 Bq/L,
^{210}Po: 0.022–0.145 Bq/L,
^{230}Th: 0.0029–0.0150 Bq/L,
^{232}Th: 0.0016–0.0056 Bq/L.

As expected, these activity concentrations are orders of magnitude below those for the gamma emitters shown in Table 1 — analyses of NORM from many different oil and gas production facilities have shown that the solids found in pipes and surface structures do not contain ^{238}U and ^{232}Th because these radionuclides are not mobilized from the reservoir rock [3]. Thus, formation water contains the radium isotopes ^{226}Ra from the ^{238}U series and ^{228}Ra and ^{224}Ra from the ^{232}Th series, but not their parent radionuclides.

3.2. Scale

The activity concentrations of ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{224}Ra and ^{40}K in the scale samples, as determined by gamma spectrometry, are shown in Table 2. The ranges of activity concentration were 38.5–58.3 Bq/g for ^{226}Ra, 0.20–0.60 Bq/g for ^{210}Pb, 26.8–39.2 Bq/g for ^{228}Ra, 8.6–15.9 Bq/g for ^{228}Th, 8.8–15.4 Bq/g for ^{224}Ra and 1.3–2.3 Bq/g for ^{40}K. These results are comparable with those obtained in other countries [3], and reflect the variations due to factors such as the geochemical and geological characteristics of the reservoir rock, the age of the well, the types of hydrocarbon produced and the operating conditions.

TABLE 2: GAMMA EMITTING RADIONUCLIDES IN SCALE

Sample code	Activity concentration Bq/g					
	Ra-226	Pb-210	Ra-228	Th-228	Ra-224	K-40
SC1	38.5 ± 0.4	0.4 ± 0.01	26.8 ± 0.1	9.8± 0.8	9.6 ± 0.9	1.8 ± 0.2
SC2	41.5 ± 0.4	0.3 ± 0.01	28.2 ± 0.5	11.0 ± 0.6	10.9 ± 0.8	1.8 ± 0.1
SC3	40.7 ± 0.4	0.2 ± 0.01	27.6 ± 0.5	10.9± 0.5	11.2 ± 0.7	1.3 ± 0.2
SC4	40.7 ± 0.1	0.3 ± 0.01	29.9 ± 0.5	8.6 ± 0.6	8.8 ± 0.6	2.0 ± 0.3
SC5	58.3 ± 0.2	0.6 ± 0.1	39.2 ± 0.7	15.9 ± 1.2	15.4 ± 0.9	2.3 ± 0.3

In the same scale samples, the activity concentrations of ^{234}U, ^{238}U, ^{210}Po, ^{230}Th and ^{232}Th were determined using alpha-particle spectrometry. The ranges for each radionuclide were as follows:

^{234}U: 0.0009–0.0045 Bq/g,
^{238}U: 0.0016–0.0046 Bq/g,
^{210}Po: 0.066–0.166 Bq/g,
^{230}Th: 0.0012–0.0039 Bq/g,
^{232}Th: 0.0009–0.0045 Bq/g.

As expected, the activity concentrations of the uranium and thorium isotopes and ^{210}Po were much lower than those of the gamma emitters shown in Table 2. As with the produced water, only the radium isotopes, and not their parent radionuclides, are leached from the reservoir rock.

4. CONCLUSIONS

Produced water and scale in the Ghanaian oilfields are similar to those found in other countries, in that they contain significant levels of NORM, particularly radium isotopes. If not properly handled and disposed of appropriately, they may become a source of concern for public exposure and the environment. This is particularly so if these materials are discarded indiscriminately without consideration of any implications for the environment. Based on the results obtained from this study, there is a need for guidance levels for radionuclides prior to discharge of effluents into water bodies in order to ensure that there is no undue contamination of the environment.

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Poster Presentation

RADIOLOGICAL ASSESSMENT OF WATER TREATMENT SYSTEMS IN POÇOS DE CALDAS, BRAZIL

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Abstract

Preliminary measurements were made of the activity concentrations of radionuclides in the ^{238}U and ^{232}Th decay series in water and process waste associated with the treatment of water in the city of Poços de Caldas, Brazil. The results indicated a need for further evaluation of the process waste in cooperation with the local water treatment company to identify any occupational and environmental radiation protection issues.

1. INTRODUCTION

Radionuclides of natural origin are found in the terrestrial biosphere, distributed throughout water bodies, biota, rocks, soil and even air [1]. When minerals are extracted from the Earth's crust and processed, the radionuclides contained within them end up in process residues, products and by-products. The industrial activities concerned include mining and minerals processing, oil extraction and water treatment. During these processes, radionuclides may become diluted or concentrated. Monitoring and management of the various process materials may therefore be important from the point of view of radiation protection of humans and the environment [2].

Radionuclides of natural origin, along with other chemical constituents, are present in water sources as a result of natural and/or anthropogenic processes. The treatment of water to remove contaminants has been identified as one of several industry sectors that might require some form of regulatory consideration [3], since it can lead to the accumulation of radionuclides in water treatment waste that might, in some situations, have implications for radiation protection [4]. The Poços de Caldas Plateau is characterized by 'radioactive anomalies' — regions with high background radiation. A large hydrographic system permeates through the anomalous areas in a region of high rainfall. These anomalies host uranium mineralization which, in the past, was exploited as the first Brazilian uranium mine. This facility, now in the decommissioning phase, is associated with waste rock piles from which acid mine drainage occurs [5, 6]. Taking all of this into account, it is likely that waste such as sludge and filter material from local water treatment plants may contain significantly elevated radionuclide concentrations [7]. For a long time, this waste has been disposed of directly into water bodies without any appropriate pretreatment [8]. This has created a need, especially in this region, for an assessment of the quality of the public water supply as well as of the waste generated in the treatment process.

2. METHOD

Feed water, treated water and waste in the form of sludge and scale were sampled in the local water treatment system for public supply. Feed water samples were collected in the feed pipes to the treatment plant and the treated water samples were collected at the exit of the process. Sludge samples were collected directly from the decanters when these were automatically drained for cleaning. Samples of scale were scraped from the walls of the decanters.

The radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb were selectively precipitated from the feed and treated water samples. The precipitates were then counted by alpha and beta techniques using a gaseous flow proportional counter Model S5-XLB Tennelec Canberra. The samples of sludge and scale were subjected to drying, milling and packaging. The determination of ^{226}Ra, ^{228}Ra and ^{210}Pb was conducted by gamma spectrometry using a Canberra Range Detector Model GX4510. The elements thorium and uranium (from both water and sludge samples) were determined by UV–visible spectrometry (Varian, model Cary 50). Thorium was extracted from aqueous solution using a solution of trioctylphosphine oxide diluted in cyclohexane, followed by re-extraction in aqueous solution. Uranium separation was carried out by extraction with tri-n-butyl phosphate in a solution containing $Al(NO_3)_3$, EDTA and tartaric acid, followed by uranium stripping with Arsenazo III.

3. RESULTS AND DISCUSSION

The results obtained for feed and treated water from two sampling campaigns at three treatment plants (A, B and C) are shown in Table 1. Screening levels of 0.5 and 1 Bq/L for total alpha and total beta activity concentrations, respectively, are established in Brazilian legislation [9]. Should these levels be exceeded, the radionuclide composition must be determined and the results compared with specified reference values which, in the case of ^{226}Ra and ^{228}Ra, are 1 and 0.1 Bq/L, respectively. It is evident from Table 1 that all the measured values for these two radionuclides fall well below the reference values. Although not included in Ref. [9], ^{210}Pb is considered important from a radiation protection point of view because it could contribute significantly to internal exposure [10]. The low U and Th concentrations found are to be expected, given the low total alpha and total beta activity concentrations.

The results obtained for sludge sampled from the same three water treatment plants are shown in Table 2. The sludge composition depends on the physical and chemical nature of the water and the types and amounts of chemicals used [11]. At Plants A and C, sediment samples were collected from around the facility for comparison purposes. While there are no regulatory criteria for sludge, the fact that the radioactivity levels in the sludge were similar to those in the sediments surrounding the plants suggests that the sludge is not of radiological concern.

The activity concentrations measured in scale samples collected during two sampling campaigns at Plants A, B and C are shown in Table 3. In the first campaign, the activity concentrations at Plants A and B were moderate, but those at Plant C were noticeably elevated. In the second sampling campaign, the concentrations at Plants A and B were again moderate, similar to those measured in the first campaign, but those at Plant C were now less elevated than before. The uranium and thorium concentrations, while all moderate, also showed differences between the first and second sampling campaigns. There is no conclusive explanation for this finding at present, and the results from further sampling campaigns are awaited with interest.

TABLE 1. RADIOACTIVITY IN WATER

Activity concentration (Bq/L)

	Each of Ra-226, Ra-228, Pb-210	U	Th	Total alpha	Total beta
First sampling campaign:					
Feed	<0.02	<0.18	<0.01	<0.09 (plant C) <0.06 (others)	<0.09 (plant C) <0.06 (others)
Treated	<0.02	<0.18	<0.01	<0.06	<0.06
Second sampling campaign:					
Feed	<0.02	<0.02 (plant A) <0.10 (plant B) <0.20 (plant C)	<0.01	<0.48	<0.65
Treated	<0.02	<0.02 (plant A <0.05 (plant B) <0.02 (plant C)	<0.01	<0.48	<0.65

TABLE 2. RADIOACTIVITY IN SLUDGE

	Activity concentration (Bq/L)				
	Ra-226	Ra-228	Pb-210	U	Th
Sludge samples:					
Plant A	198 ± 50	179 ± 9	145 ± 7	211.19 ± 7.92	214.92 ± 23.46
Plant B	184 ± 46	156 ± 8	151 ± 8	475.30 ±3.03	130.06 ± 14.56
Plant C	183 ± 46	174 ± 9	209 ±1	702.83 ± 8.88	210 ± 21
Sediment samples from around the plant (for comparison):					
Plant A	110	226	144	161	228
Plant C	214	293	350	379	298

TABLE 3. RADIOACTIVITY IN SCALE

	Activity concentration (Bq/L)				
	Ra-226	Ra-228	Pb-210	U	Th
First sampling campaign:					
Plant A	69 ± 17	71 ± 4	37 ± 2	<505.63	<202.2
Plant B	164 ± 41	226 ± 11	30 ± 2	<505.63	<202.2
Plant C	1192 ± 298	1704 ± 85	301 ± 15	<505.63	<202.2
Second sampling campaign:					
Plant A	69 ± 23	105.4 ± 9.5	66 ± 20	<244	134
Plant B	164 ± 41	252 ± 18	74 ± 19	<244	<103
Plant C	619 ± 110	790 ± 52	98 ± 24	<244	170

4. CONCLUSIONS

From these preliminary results, it can be concluded that the levels of natural radioactivity in the water provided to the population of Poços de Caldas municipality do not pose any concern for health. The levels of radioactivity in the sludge waste from the water treatment process are similar to those found in sediments in the surroundings of the water treatment plants and no health or environmental concern is indicated. Preliminary results for the scale precipitated in the treatment tanks show that it contains moderately elevated concentrations of Ra and Pb isotopes. Some differences between the results from the first and second sampling campaigns were found. It is hoped that further sampling campaigns will provide some explanation.

It is known that the region has well defined climatic situations, as well as an intense precipitation period. The intention is to continue the study over the course of a year to investigate any influence of seasonal effects. Once the study is complete, the presence of radionuclides in the scales of the treatment tanks will be evaluated regarding possible occupational and environmental radiation protection issues, in cooperation with the local water treatment operator.

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Poster Presentation

BACTERIAL LEACHING IN URANIUM MINE EFFLUENTS AT THE ORE TREATMENT UNIT, CALDAS, BRAZIL

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Abstract

The condition for bacterial leaching (*Acidithiobacillus* spp.) in uranium ores is the presence of metallic sulphides such as associated pyrite (FeS_2), which occurs in the ore and waste rock piles of the Uranium Ore Treatment Unit (UTM), Minas Gerais State, Brazil. In the UTM area, water sampling was conducted at three points in the UTM interface environment, at two other points in the region (the open pit mine and waste rock piles) and at two points in the tailings dam. Chemical and bioleaching bacteria density analyses were performed using each sample. Water samples from all sampling points at the UTM interface, namely Points 25, 76 and 41, presented mean values of manganese concentration (0.10 mg/L) and fluoride (1.40 mg/L) which exceeded the limits established by Brazilian legislation. The mean value of uranium concentration found at Point 76 (0.13 mg/L) was also above the limit established by Brazilian legislation. Water samples from P76 (UTM interface) were considered susceptible to acid mine drainage and to the activity of bacteria involved in the bioleaching of metals. The chemical and microbiological results obtained in this study support the need to review actions currently employed by UTM for effluent control.

1. INTRODUCTION

Sulphate minerals present in mining areas may cause serious environmental problems due to chemolithotrophic bacterial action of the genus *Acidithiobacillus*, mainly *A. ferrooxidans* and *A. thiooxidans*. These microorganisms are capable of oxidizing mineral sulphates, elementary sulphur and the ferrous ion (*A. ferrooxidans*), and of mobilizing metals such as uranium to the environment [1, 2]. The condition for bacterial leaching (*Acidithiobacillus* spp.) in uranium ores is the presence of metallic sulphides such as associated pyrite (FeS_2), which occurs in the ore and waste rock piles of the Uranium Ore Treatment Unit (UTM), Minas Gerais State, Brazil. Legal procedures for remediation of the area and facility decommissioning are currently underway [3]. In this context, the main goals of this study were: (i) to evaluate the ecologies of populations of the bioleaching microorganisms (*A. ferrooxidans* and *A. thiooxidans*) in effluents from mining companies containing sulphides and associated radionuclides, and (ii) to assess the physicochemical and radiological uranium mining impact on surface water quality.

2. MATERIALS AND METHODS

2.1. Sampling

Quarterly sampling was carried out between October 2008 and July 2009. Water was collected in 5 L Van Dorn bottles, transferred to previously washed plastic bottles and stored at 4°C prior to analysis. In the area of the UTM, three points in the UTM interface environment (Points 25, 41, and 76), three points in the open pit mine and waste rock piles (Points PM, 75 and D3) and two points in the tailings dam (Points 27 and 32) were sampled, as shown in Fig. 1.

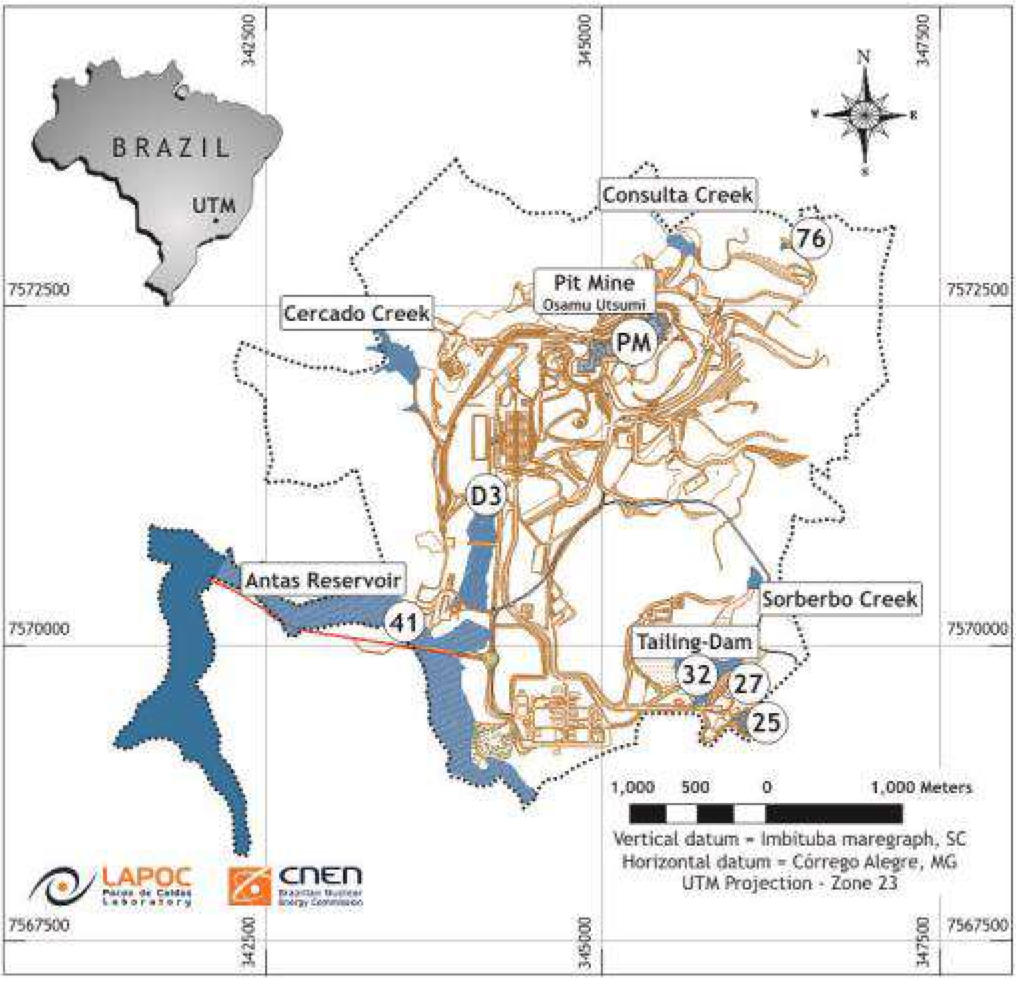

FIG.1. UTM sampling points (drawing by H.L.C. Albertini and E.O. Lima-Filho).

2.2. Chemical measurements

The pH values were measured using a pH meter (model DM-21, Digimed, São Paulo) with a selective electrode. The analyses of hardness (carried out by measuring the concentration of Ca and Mg) and manganese concentrations were conducted using coupled plasma atomic absorption spectrometry. The fluoride concentration was measured using a potentiometer with selective electrode [4, 5]. Uranium concentrations were measured through spectrophotometry with Arsenazo III as the reagent [6].

2.3. Microbiological measurements

The three tube ‘most probable number’ technique was used to estimate the population density of *A. ferrooxidans* and *A. thiooxidans*. A series of adequate dilutions (1:10), made of each sample using sterilized distilled water at pH = 2.0, was carried out to determine the occurrence of populations of *A. ferrooxidans* and *A. thiooxidans*. Aliquots of 1 mL from each dilution were then transferred to sampling tubes containing 10 mL of T and K medium with ferrous ion (Fe^{2+}) as a source of energy, and to tubes containing the same basal medium with elemental sulphur as the source of energy for detection of *A. ferrooxidans* and *A. thiooxidans*, respectively [7]. The prepared samples were incubated in a 200 rpm shaker at 30°C for 21 d.

2.4. Statistical analysis

The results of the physical and chemical analyses were evaluated using ANOVA and Tukey’s test (post hoc test) to detect significant differences between water samples obtained from the different sites. The statistical tests above were performed using the BioEstat 4.0 program [8]. Pearson’s correlation coefficient (r) was used to find correlations between the population densities of *A. ferrooxidans* and *A. thiooxidans* and the values of chemical variables obtained for the water samples [9].

3. RESULTS AND DISCUSSION

The average pH values ranged from 3.45 (Point D3) to 9.61 (Point 32). The average pH value recorded in water samples at Point 32 (9.61) was evidently higher ($P<0.05$) than the average values recorded in water samples from the other points. The pH values observed at Point PM (3.84), Point D3 (3.45) and Point 75 (3.68) were similar ($P>0.05$). Average pH values in water samples from Point 27 (4.80) and Point 76 (4.97) did not present differences between them ($P>0.05$).

The average hardness value observed in water samples from Point 76 (17.95 mg/L) was lower ($P<0.05$) than the average value obtained from samples at Point PM (927.05 mg/L) and the average concentration obtained at Point 75 (256.07 mg/L). The average hardness value from water samples from Point 25 was 535.65 mg/L.

The greatest average fluoride value was registered in water samples from Point 75 (132.6 mg/L) and it was higher ($P<0.05$) than all other registered values at UTM. Average values of fluoride in water samples from Point PM (55.32 mg/L) and Point D3 (19.55 mg/L) were similar to each other ($P>0.05$) and higher ($P<0.05$) than values recorded at Points 32, 27, 25 and 76. Average concentrations recorded at Points 32 (2.64 mg/L), 27 (9.84 mg/L), 25 (2.42 mg/L) and 76 (2.09 mg/L) did not differ significantly from each other ($P>0.05$). The fluoride values in samples from Points 25 and 76 exceeded the limit for Class 2 water bodies, according to CONAMA Resolution 357 [10].

High average uranium concentrations were observed in water samples from Point PM (2.94 mg/L), Point D3 (5.20 mg/L) and Point 32 (5.85 mg/L). The average value of uranium obtained at Point 76 (0.13 mg/L) exceeds the maximum limit established by CONAMA Resolution 357 for Class 2 water bodies [10] and by document CNEN No. 50/SLC, [11].

Water samples from all sampling points at the UTM interface, i.e. Points 25, 76 and 41, presented manganese concentration values above the limit established for Class 2 water bodies (0.10 mg/L) [10].

At Point 41, at the UTM interface with the environment, the average pH value observed was 6.60 while the average hardness value was 122.5 mg/L. Both the observed average fluoride value of 1.89 mg/L, as well as the average value of manganese (0.66 mg/L) recorded at Point 41, exceed the limits established by CONAMA 430 [12]. Results registered at sampling points at the UTM interface (Points 25, 76 and 41) are in agreement with Ref. [13] and support the need for reviewing actions currently taken by UTM regarding effluents. It has been recognized that bioavailability and toxicity of metals in water are dependent on chemical variables such as pH and hardness [14, 15]. The results of this study agreed with Ref. [16], and since Point 76 was categorized as soft (average hardness value of 17.95 mg/L) and acidic (average pH value of 4.97), higher bioavailability of metals in that environment could be expected.

The highest average values of *A. ferrooxidans* were detected in water samples from the open pit mine and waste rock piles points, which are Points PM (3775 NMP/mL), D3 (5375 NMP/mL) and 75 (14700 NMP/mL). The highest average value of *A. thiooxidans* (7325 NMP/mL) was detected at Point D3, while the lowest average value (12 NMP/mL) was observed at Point 41. Among samples from the tailings dam region, those from Point 27 presented the highest average values of *A. ferrooxidans* (3600 NMP/mL) and *A. thiooxidans* (1200 NMP/mL).

The lowest average values of *Acidithiobacillus* spp. were recorded in samples from the UTM interface with the environment, which are Points 25 (25 NMP/mL), 41 (307 NMP/mL) and 13 (489 NMP/mL). Among samples collected at the UTM interface, those from Point 76 presented the highest average occurrence value of *A. ferroxidans* (378 NMP/mL), followed by the second highest average value of *A. thiooxidans* (112 NMP/mL). According to the results presented, the lowest average values of *A. ferrooxidans* and *A. thiooxidans* were detected in a water sample collected at Point 25 (12 MPN/ml) and this result may be possibly related to the high average pH value detected (7.26). These results are confirmed by the negative correlation (r = –0.80) verified between the bacteria concentrations and the pH value found at the mentioned point. On the other hand, the highest average values of acidophilic bacteria were detected in water samples from Points PM, D3 and 75 (open pit mine and waste rock piles), where the lowest average pH values were registered. Such result was confirmed by the negative correlation verified between the average pH value and the density of *A. thiooxidans* (r = –0.87). The results obtained comply with what is expected, since the optimal pH value for these species growth is approximately 2.0, although growth is observed in the range 1.2–4.0 [17].

The populations of *T. ferrooxidans and T. thiooxidans* presented remarkable seasonal variation, both quantitatively and in relation to their incidence in the different evaluated sampling points. Within the sampling points at the UTM interface with the environment, Point 76 was considered susceptible to acid mine drainage and to the activity of bacteria involved in metals bioleaching. The chemical and microbiological results obtained in this study support the need to review the actions currently taken by UTM for effluent control.

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INDUSTRY SPECIFIC STUDIES
(e.g. OIL, GAS, COAL, METALS)

(Session 6)

Chairperson

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Brazil

PRACTICAL CASES OF NORM TRANSPORT: PROBLEMS AND SOLUTIONS

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Abstract

A serious and complex issue in the transport of NORM is associated with international NORM shipments. This process requires involvement of several logistics companies and government departments in different countries, which typically do not have a full understanding of the legislative requirements and associated hazards, or sometimes are not aware of them at all. The following practical examples are discussed:

(a) Transport of mixed dangerous goods where the presence of other hazardous substances contained in the material must also be considered,
(b) Application of regulations to NORM containing radionuclides that are not in secular equilibrium, and to material at transit locations,
(c) Detection of NORM at border crossings,
(d) Buildup of radon in containers and hulls of ships,
(e) Selection of correct surface contamination limits,
(f) The lack of communication in international trade in NORM.

1. INTRODUCTION

There are many situations where — in the handling, storage and transport of Class 7 (radioactive) dangerous goods — other hazardous substances contained in the same material also have to be considered. An additional issue associated with the transport of Class 7 dangerous goods (especially where other hazardous substances are present in the material) is the general ignorance of both the legislative requirements and of potential exposures of the transport workers to hazardous substances. The situation becomes much more complex when the material is transported internationally, involving several companies and government departments in different countries that typically do not have a full understanding of the legislative requirements and associated hazards, or are not aware of them at all. This issue has already caused:

- Several mineral shipments to be returned to the countries of origin or to be held for a long time at customs, resulting in financial difficulties for the producers and logistics companies;
- Workers compensation and other successful legal claims for the injury caused by an unknown level of exposure to a hazardous substance, and for the diminution of the values of the properties located in the vicinity of the transport routes.

2. TRANSPORT OF MIXED DANGEROUS GOODS

The International Maritime Dangerous Goods Code [1] and Australian Dangerous Goods Code [2] contain the following requirement:

> “5.1.4 Mixed packing
> When two or more dangerous goods are packed within the same outer packaging, the package shall be labelled and marked as required for each substance.”

In many cases this requirement remains unknown to the personnel of logistics companies and relevant government departments, and is not being followed as required. The following five cases describe the situation where mixed dangerous goods were transported in accordance with the legislative requirements. For reasons of confidentiality, companies are not named.

2.1. Radioactive and environtally hazardous material — Africa

A specific addition was made to DG Class 9: “Environmentally Hazardous Substances NOS”. In many cases in accordance with the requirement 5.1.4 on mixed packing in addition to any other DG label, an additional one may need to be placed on the package. The transport of uranium concentrate from an African country to an overseas customer is an example of full compliance with the IMDG Code [1], where each drum with the concentrate and the container in which these drums are placed are labelled as both a “radioactive” and “environmentally hazardous” substance.

2.2. Radioactive and corrosive material — Australia

In the process of remediation of a site in Australia, several corroded drums with sludge from mineral processing were discovered. The material needed to to be transported on a public road for reprocessing. It was known that it was radioactive and needed to be labelled as such for transport. However, taking into account the poor condition of the drums and the information that the sludge was originally highly acidic (pH of the order of 1) the drums were placed into a lined container and labelled as both “radioactive” and “corrosive”.

2.3. Radioactive and biologically hazardous material — Asia

At a water treatment plant in Asia, the sludge is transported for disposal in small trucks. The material was considered to be a biologically hazardous substance until relatively high concentrations of ^{226}Ra were found in the sludge. The truck now bears two labels, both “biologically hazardous” and “radioactive”. In this case it was considered that labels in a local language are preferable.

2.4. Radioactive and flammable material — Middle East

In a country in the Middle East, radioactive sludge from oil and gas production is being placed into drums for eventual disposal in an approved facility. When the drums are transported to the disposal sites they are labelled as both “radioactive” and “flammable”. Similarly to case 2.3 above, it was considered that labels in a local language are more useful.

2.5. Radioactive and toxic material — Middle East

Pipes from oil and gas production facilities are transported to a NORM processing facility where internal scale is removed and the pipes are subsequently returned to service. Owing to the radium bearing scales being on the inside surfaces, the pipes were plugged at both ends and the vehicles were labelled as transporting SCO (surface contaminated material). When safety personnel at the processing facility discovered that pipes coming from one oil field also contained relatively high concentrations of mercury, additional safety procedures were introduced at the plant and vehicles transporting the pipes are now also labelled with a “toxic” sign. As in the case above, labels in the local language, from the locally applicable Dangerous Goods Code, were placed on the vehicles.

3. APPLICATION OF REGULATIONS TO MATERIALS IN WHICH RADIONUCLIDES OF NATURAL ORIGIN ARE NOT IN SECULAR EQUILIBRIUM

To ensure that material is transported correctly, two IAEA documents need to be consulted: the Transport Regulations [3] and the Advisory Material [4]. The ‘factor of 10’ used to assess the applicability of the Transport Regulations is well known, but the case where radionuclides are not in a state of secular equilibrium is not fully understood by many companies and government departments. Paragraph 107 (f) of the Transport Regulations [3] states:

> “For natural materials and ores containing naturally occurring radionuclides that are not in secular equilibrium the calculation of the activity concentration shall be performed in accordance with para.405.”

The Advisory Material [4] provides additional information for these cases:

> “…the basic nuclide values for exempt activity concentration as given in Table 2 for U(nat) and Th(nat) can only be used if the radionuclides are in secular equilibrium. If this is not the case, owing to processing activities such as chemical leaching or thermal treatment, the natural radioactive equilibrium state does not exist and the formula for mixtures of radionuclides according to para. 405 has to be applied to calculate the exempt activity concentration.”

Complete data on the possible disruption of secular equilibrium in the ^{238}U and ^{232}Th decay chains during processing of mineral concentrates is not available in many cases, and it is prudent to assume that this may occur in case of:

(a) Any chemical processing of the material, such as leaching or adding flotation agents;
(b) Any thermal processing of the material — owing to the variety of materials it is impossible to establish a universal cut-off point for the temperature at which some radionuclides (such as ^{210}Pb and ^{210}Po) could volatilize and disrupt the equilibrium but a value of 250–300°C is suggested as a general guide at which additional analysis of the material may be required;
(c) Any combination of chemical and thermal treatment of ores and minerals.

The data would typically be required for ^{238}U, ^{230}Th, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra and ^{228}Th as appropriate. For example, in the heavy mineral sands industry (titanium and zirconium minerals):

(a) The mineral concentrates (separated using gravimetric methods) and individual minerals (separated using electrostatic and electromagnetic methods) are analysed for thorium and uranium only, as the separation process does not disrupt the secular equilibrium in the ^{238}U and ^{232}Th decay chains.
(b) The minerals that have undergone (i) chemical treatment (such as washing zircon sand grains with acid solution), (ii) thermal treatment (such as heating titanium mineral ilmenite to remove excessive iron in the production of synthetic rutile) and (iii) the combination of chemical and thermal treatment (further treatment of synthetic rutile to remove other impurities) should be analysed for other radionuclides in the ^{238}U and ^{232}Th decay chains, prior to the decision on the applicability of Transport Regulations [3] to these materials.

(c) All materials in the downstream processing of heavy mineral sands (e.g. in the production of titanium dioxide pigment and fused zirconia) should also be analysed for other radionuclides in the ^{238}U and ^{232}Th decay chains.

4. APPLICATION OF THE TRANSPORT REGULATIONS TO MATERIALS IN A TRANSIT STORAGE AREA

In some jurisdictions, specific provisions have been made for material that is located at a transit storage area. The main reason for such a provision is that, typically, mining and mineral processing companies transport their products not directly to the port but to a transit location, where a sufficient number of containers or bulk material would be accumulated prior to their transport to the nearby port. Thus, a certain volume of material is almost always present at a transit location. This results in a situation where, from one point of view, the material could be considered to be ‘in transport’ but, from the other point of view, the material could be considered to be ‘in ‘ storage’. In the latter case, separate regulatory control over that storage may be needed. In order to address this situation, different arrangements could be made. In Western Australia, for instance, if NORM containing U(nat) and Th(nat) in concentrations between 1 and 10 Bq/g[1]) is stored at any transit location for more than 24 h, regulation 28 of the Radiation Safety Regulations [5] (specifying conditions on the registration of premises) would apply and the transit location would have to be registered for storage of radioactive substances with the appropriate authority, namely the Radiological Council of Western Australia.

5. DETECTION OF GAMMA RADIATION FROM NORM PACKAGES AT BORDER CROSSINGS

Irrespective of whether a consignment of NORM is exempt from the Transport Regulations and the associated labelling requirements, the concentrations of radionuclides in the package (e.g. a shipping container) may give rise to gamma dose rates outside the package that are easily detectable by the equipment that is commonly used at border crossings and in ports worldwide. The triggering of portal alarms at border crossings causes significant operational issues, as all such alarms should be fully investigated. After a shipment of NORM has triggered an alarm, the resulting investigation would include the identification of the relevant radionuclides, interviews with the personnel involved and an examination of all relevant documentation. Therefore, the transport documentation for many materials needs to contain detailed information about the concentrations of naturally occurring radionuclides in this material, irrespective of its classification. As the requirements for this documentation differ from country to country, all necessary information may be provided in the Material Safety Data Sheet (MSDS), the document that accompanies every material shipment. The inclusion of the gamma spectrum for a particular material into the MSDS, in the form of either a table or a chart, is highly advisable. While not absolutely necessary, this information would assist in the process of clearing a particular NORM through the radiation detection equipment at international border crossings.

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[1] In terms of Table 2 and para. 107(f) of the Transport Regulations [3], the values of 1 and 10 Bq/g are, respectively, the exemption level and 10 times that exemption level applicable to this type of NORM.

6. BUILDUP OF RADON IN CONTAINERS AND HULLS OF SHIPS

Irrespective of whether a material is exempt from the Transport Regulations and the associated labelling requirements, the concentrations of ^{238}U and/or its decay progeny such as ^{226}Ra may give rise to a significant buildup of radon (^{222}Rn) inside sealed shipping containers and hulls of ships used for the transport of bulk minerals. Where such an occurrence is discovered, the typical approach is to instruct workers opening containers and ships' hulls at the destination to stay away from the material for a certain time (typically 1 h) to allow for radon concentrations to decrease through natural ventilation.

In Australia, for example, when a material containing only 1.5 Bq/g of ^{238}U was stored in a sealed container, the concentratetion of ^{222}Rn reached 8000 Bq/m^3 after approximately 36 h. It was highly noteworthy that a worker dealing with 'exempt' material such as this would have received an inhalation dose of 1 mSv in just over 22 h and 20 mSv in about 450 h.

7. SELECTION OF CORRECT SURFACE CONTAMINATION LIMITS

The Transport Regulations [3] provide the following definitions for 'surface contamination' and 'low toxicity alpha emitters:

> "214. Contamination shall mean the presence of a radioactive substance on a surface in quantities in excess of 0.4 Bq/cm^2 for beta and gamma emitters and low toxicity alpha emitters, or 0.04 Bq/cm^2 for all other alpha emitters.
>
> …
>
> "227. Low toxicity alpha emitters are: natural uranium, depleted uranium, natural thorium, uranium-235, uranium-238, thorium-232, thorium-228 and thorium-230 when contained in ores or physical and chemical concentrates; or alpha emitters with a half-life of less than 10 days."

Typically, the radionuclides associated with NORM contamination on the surfaces of objects in transport will, with the notable exception of ^{226}Ra, comprise only low toxicity alpha-emitters. The fact that ^{226}Ra is not classified as a low toxicity alpha emitter is often not appreciated or is overlooked. Consequently, while the surface contamination limit of 0.4 Bq/cm^2 generally applies to all NORM surface contaminated objects in transport, in specific situations (such as when transporting contaminated objects from the oil and gas industry or from some plants for the production of titanium dioxide pigment), the limit of 0.04 Bq/cm^2 is applicable instead.

8. LACK OF COMMUNICATION IN INTERNATIONAL TRADE INVOLVING NORM

In many cases, there is an insufficient level of communication with regard to the legal requirements between the exporter and importer of a mineral concentrate containing NORM. The lack of communication between logistics companies and government departments in different countries on this issue and misinterpretations of different legislation, regulations and guidelines often results in serious problems, particularly for the customers of the mining and mineral processing industry. The issue becomes much more complex if we consider the fact that Transport Regulations [3] are not adopted uniformly across the world and different requirements may apply in different jurisdictions. Each of the examples below presents a situation that has been encountered in different countries in the process of the import of NORM and it is hoped that these issues will no longer arise for the mining and mineral processing industry.

8.1. Containers held in port owing to lack of documentation

Three examples are given here of where problems have occurred during the process of importing NORM in containers — in all three cases, the material was not classified as radioactive in terms of the latest edition of the Transport Regulations [3]:

(i) In the first example, a portal monitor alarm was triggered by containers at the border control point. The resulting situation became aggravated by the following factors:
   - Only basic information on radionuclide concentrations was available in the MSDS;
   - The portal monitors had been installed only a few days prior to this event and this was the first instance of the alarm being triggered;
   - A heated argument ensued between a customs official and the shipping agent — the agent was demonstrating documents in English, a language that the border control personnel did not understand.

   The shipping agent was arrested and spent several days in prison for "trying to import radioactive material into the country illegally". The containers were held in a quarantined area of the port for several months.

(ii) In the second example, containers with mineral concentrate sent from a country where an earlier version of the Transport Regulations was in force were refused entry into another country where the latest version of the Transport Regulations [3] was in force. In terms of the earlier version of the Transport Regulations, the composition of the material had to be analysed only for uranium and thorium whereas, in terms of the current version, other radionuclides had to be included in the analysis. The containers were held at the port for several weeks and were only released after additional analyses of the material were carried out and the relevant data were made available.

(iii) In the third example, the opposite situation occurred. Containers with mineral concentrate from the country that had adopted the latest version of Transport Regulations (the 2005 edition at that time) were transported to a country where the local transport regulations had not been updated since 1987. This resulted in a situation where the NORM concentrate was, for purposes of transport, exempt in the country of origin (and internationally) but had to be classified and labelled as 'radioactive' in the importing country.

8.2. Country- and port-specific guidelines and standards

There may be a guideline applicable in a country (or even in one particular port) that is not known to the exporting company. An example of such a guideline is a standard that was in force in China in 2005 that specifically addressed the inspection of the radioactivity content during the importation of minerals [6]. The procedure set out in that standard was based on a comparison of the background radiation level and the radiation emitted from a particular material. If gamma dose rates measured on the surface of the imported material exceeded a predetermined value (or were more than ten times higher than the background gamma radiation), unloading of the material had to be stopped immediately, pending additional investigations and sampling. In some cases the entry of the mineral concentrate into the country was refused and the mineral had to be shipped back to the country of origin or to another potential customer. Several other cases are known where mineral concentrates were similarly refused entry to a country, resulting in significant financial and commercial difficulties for the exporter, importer and logistics companies involved in the transport of this type of NORM.

8.3. Transit of NORM shipments through international ports

Many NORM concentrates that are transported in containers are transshipped through various international ports, where the containers are transferred to other vessels. It is important to note the following:

(a) In some ports, the 'radiological screening' of containers (as discussed in Section 5) would still have to take place. In such cases, it is advisable for the logistics companies and relevant shipping agents to personally visit the port, explain the characteristics of the material and present samples of the material to the Port Chemist. This process has been carried out several times by different Australian companies and has eliminated any issues with certain types of NORM being transshipped through one of the Asian ports.
(b) In other ports, both import and export licences may be required, even if a container with a mineral concentrate only stays in a port for a day or two. In such cases, it would be essential to involve a locally registered shipping agent to obtain the necessary import and export licences. In one instance, the material received only an import licence, whereupon further transshipment to the destination was delayed for a considerable time while an export licence was obtained.

9. THREE COMMON MISTAKES

In addition to the communication-related problems exemplified in Section 8, three common mistakes made in the transport NORM can be identified:

(1) In the transport of mineral exploration samples, an exploration company usually does not have data on radionuclide concentrations and, because the application of the Transport Regulations to the transport of exploration samples is not widely understood, the shipments are usually not labelled in any way. In many of these cases, the samples may be transported as an 'excepted package' in accordance with para. 516 of the Transport Regulations [3], which states: "The radiation level at any point on the external surface of an excepted package shall not exceed 5 microSv/h".
(2) At many mining and mineral processing sites, the individuals responsible for the transport of NORM receive data on the concentrations of thorium and uranium from an on-site laboratory. A common mistake made in calculating the activity concentrations of thorium and uranium is that a typical laboratory would provide the data in units of parts per million (or µg/kg) for ThO_2 and U_3O_8, rather than for Th and U. In this regard, it is important to note that 1 ppm of ThO_2 is equivalent to 0.879 ppm of Th, while 1 ppm of U_3O_8 is equivalent to 0.848 ppm of U.
(3) When mineral concentrates are transported in bulk, relatively often the vehicles (especially trucks with trailers) are not labelled appropriately and the Transport Index is not adjusted accordingly. It is essential to ensure that, in accordance with para. 248 and Table 7 of the Transport Regulations [3]:
    - The definition of the vehicle is used when considering the labelling of the vehicles;
    - The multiplication factors for unpackaged LSA-I and SCO-I associated with the "largest cross sectional area of the load being measured" are taken into account when calculating the Transport Index for the shipment.

10. ASSOCIATED LEGAL ISSUES

In some situations companies and government departments may become involved in legal challenges without actually transporting radioactive material or exposing workers and/or the general public to any levels of radiation. The fear of radiation has been described in detail in Refs [7, 8] and many other papers and documents. Unfortunately, this fear continues to prevail, resulting in shipping companies generally being unwilling to transport any substance that is labelled “radioactive”. Even if material is transported as an “excepted package” [3], when the warning labels need to be present only inside the shipping container, the fact that the sign “radioactive” must be visible when the container is opened may create an unwarranted panic in case of an accident. The following cases describe two situations where a legal challenge was successful, despite the fact that no radiation exposures had actually taken place.

10.1. Diminution of property values

In the United States of America, the perceived risk of radiation exposure was determined to be a reason for litigation and subsequent compensation in the case of New Mexico Supreme Court in Santa Fe v. Komis, 845 P.2d 753 (1992), which addressed a claim of diminution of property values based on perceived risks of nuclear waste transportation. The case involved partial condemnation of land taken by the City of Santa Fe to construct a highway for primarily normal public use but also for occasional transportation of nuclear waste from Los Alamos to the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. The jury in the District Court awarded $489 582 for the land actually taken, $60 784 for severance damages to the buffer zone and $337 915 for perceived loss due to public perception. The New Mexico Supreme Court affirmed judgement of the District Court and stated: “If people will not purchase property because they fear living or working on or near a WIPP route, or if a buyer can be found, but only at a reduced price, a loss of value exists. If this loss can be proven to the jury, the landowner should be compensated” [9].

10.2. Compensatable injury from fear of radiation

Another court case from the United States of America [10], which was reported at the NORMV International Symposium [11], indicates that a person could sustain what was described as a “compensable injury” simply from fear of radiation. This particular case was a result of a truck driver’s contact with a leaking container that was mistakenly labelled as radioactive waste. Although the driver suffered no physical injuries and was not actually exposed to radiation, the court determined that the driver’s post-traumatic stress disorder, depression, fatigue and anxiety were rationally connected to his contact with the hazardous material and were therefore compensatable under Tennessee Law.

11. CONCLUSIONS

(i) The transport of mixed dangerous goods is, almost always, a very complex issue. It is hoped that the examples in this paper will be useful both to the companies involved in this process and to government departments administering the relevant transport safety regulations.

(ii) It is expected that specialist advice will be required in many cases to ensure compliance with all relevant regulations and guidelines.

(iii) Mining and mineral processing companies, logistics and shipping companies, and relevant government departments may need such advice in the absence of qualified personnel familiar with all requirements. An IAEA publication is available for this process [12], It is aimed primarily at government authorities but relevant companies and organizations may also find it useful to adopt the general principles contained therein.

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NORM RELATED MINERAL DEVELOPMENTS IN FINLAND

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Abstract

Many mineral resources have complex, polymetallic assemblages, in which gold, base metals, rare earth elements, niobium or phosphates are associated with NORM. Although there are no primary uranium deposits being mined in Finland, there are a number of polymetallic mineral resources and mines at various stages of assessment and development, in which uranium occurs at elevated concentrations; these include the Talvivaara nickel mine in Sotkamo, the Juomasuo and Hangaslampi gold deposits in the Kuusamo region, the Sokli phosphate mine project in eastern Lapland, the Katajakangas Nb–rare earths deposit in the Otanmäki area, and the Rompas gold–uranium prospect in Ylitornio. Detailed mineralogical characterization of these ore deposits is essential for understanding the deportment of radionuclides during mining and mineral processing, in order to both optimize metal recovery and ensure compliance with environmental guidelines. The paper briefly describes NORM related developments in the mining industry including the operating Talvivaara mine and recent exploration projects.

1. INTRODUCTION

The bedrock of Finland consists almost entirely of Archean and Proterozoic rocks of the Fennoscandian Shield. There has been a steady increase in mineral exploration and mine development in Finland over the last two decades and currently 45 mines are in operation. Of these, the majority exploit carbonate rocks (marbles) and industrial minerals, while deposits are mined primarily for metallic commodities, the most significant being nickel, zinc, copper, chromium and gold. Many mineral resources are of relatively low grade, or have complex, polymetallic assemblages, in which gold, base metals, phosphates and rare earth elements (REE) may also be associated with NORM; the latter association reflects the fact that REE, uranium, and thorium exhibit similar geochemical behaviour in rock-forming magmatic processes. Accordingly, mining and mineral processing of NORM bearing ores can lead to elevated concentrations of uranium and thorium, and their progeny (e.g. ^{226}Ra, ^{222}Rn, ^{210}Pb, ^{210}Po) in minewater, tailings, waste rock and mineral dust, resulting in potential radiation and chemical risks.

The average uranium and thorium concentrations of Finnish bedrock are 2.0 ppm and 8.9 ppm, respectively [1]. There is currently no uranium production in Finland, and known uranium deposits are of relatively low grade, small and uneconomic for exploitation. However, several mine development and exploration projects in Finland are currently focused on uranium bearing polymetallic mineral resources, the most important being the Talvivaara nickel mine, several gold deposits in the Kuusamo region, the Sokli phosphate mine project and the Rompas gold–uranium prospect. Some uraniferous polymetallic deposits also have potential for recovery of uranium as a by-product. The mineralogy of uranium and thorium has a significant influence on the deportment of radionuclides of natural origin during mining and mineral processing. Therefore, thorough mineralogical characterization and processing tests of uranium bearing and thorium bearing ores should be carried out prior to mining activities, to define the deportment of radionuclides during mineral processing, and to determine which process fractions contain NORM.

2. RECENT NORM RELATED MINERAL DEVELOPMENTS IN FINLAND

2.1. Talvivaara black schist hosted Ni–Zn–Cu–Co deposit

2.1.1. Geological environment, ore characteristics and uranium mineralogy

The Talvivaara Ni–Zn–Cu–Co deposit is hosted by metamorphosed black shales in the Kainuu schist belt, eastern Finland. It is a low grade, large tonnage deposit averaging 0.23 wt% Ni, 0.50 wt% Zn, 0.13 wt% Cu, 0.0172 wt% Co and 0.0017 wt% U [2]. The main minerals in the Talvivaara ore are quartz, microcline, anorthite, phlogopitic biotite, muscovite, graphite, pyrite and pyrrhotite. Nickel is mostly incorporated in pyrrhotite and pentlandite, zinc in sphalerite, copper in chalcopyrite, and cobalt in pentlandite and pyrite [3]. A significant part of uranium at Talvivaara is incorporated in uraninite (UO_2), which also contains thorium and radiogenic lead. Uraninite (with particle sizes of 5–100 mm) is typically enclosed in poorly crystalline, undeformed and globular carbonaceous nodules. In addition to uraninite, a small proportion of uranium is incorporated in metamorphic phosphate minerals, monazite and xenotime [4].

2.1.2. Exploitation

Production from the Talvivaara ore deposit commenced in 2008. The production process includes open pit mining, crushing, heap leaching, metals recovery and removal of metals having no current value. The leach solution percolates to the bottom of the leach pads and is either recirculated through the heap or is fed to the metals recovery section. The acidity of the leach solution (pH 2.0–2.5) is controlled by sulphuric acid, and leaching of metals is catalysed by bacteria endemic to the Talvivaara area (e.g. *Acidithiobacillus ferrooxidans*) [5]. After two years of primary leaching, the ore is reclaimed and restacked for secondary leaching to enhance leaching of metals. The secondary leaching heaps also constitute the final disposal sites for the leached ore. During metals recovery, copper, zinc, nickel and cobalt are precipitated from the pregnant leach solution (PLS) and filtered to produce saleable metal products. After the target metals have been recovered, the solution is further purified to remove unwanted metals and returned to irrigate the heaps. During removal of residual metals, the pH of the PLS is raised to 9–10, using a lime slurry, leading to precipitation of residual metals (Mn, Mg, Fe) as hydroxides, together with gypsum. The resulting slurry is thickened and the thickener underflow is directed to gypsum waste ponds.

The Talvivaara ore deposit contains about 22 000 t of uanium in 1305 Mt of measured and indicated resources grading 0.0017% U. In 2010, Talvivaara announced plans to recover uranium as a by-product. During 2011–2013, the uranium solvent extraction plant was built as a new unit in the metals recovery complex of Talvivaara. However, approval of the licensing process for uranium production is still pending, and there is no precise estimate for the anticipated start of uranium recovery at Talvivaara.

2.1.3. *The deportment of uranium and its progeny in the mining process*

A large proportion of the uranium in the black schist ore dissolves in the acidic PLS during heap leaching. Most of the uraninite is dissolved during the first half-year of the leaching [6]. The acidic and oxidative conditions of heap leaching are favourable for oxidative dissolution of uraninite, achieved especially by the oxidant Fe^{3+}. In the heaps, the oxidation of iron sulphides generates ferric iron (Fe^{3+}) which oxidizes U^{4+} to U^{6+}, leading to uranium dissolution in the PLS. The oxidation of uraninite generates dissolved uranyl $(U^{6+}O_2)^{2+}$ oxyanions and Fe^{2+}. This Fe^{2+} can then be oxidized to Fe^{3+}, which in turn oxidizes uraninite, producing more Fe^{2+}, and so on [7]. The uranium concentration in the PLS ranges between 15-28 mg/L U [6].

Uranium mostly ends up in the gypsum pond wastes and partly in the Ni–Co sulphide concentrate product; the latter is consigned to the Norilsk nickel refinery at Harjavalta [8]. Uranium residuals are extracted from the Ni–Co concentrate at Harjavalta, and reported to the Radiation and Nuclear Safety Authority (STUK). The uranium concentration of the gypsum pond wastes is highly variable (6–260 ppm U), being about 100 ppm U on average [6]. Uranium in the gypsum pond waste is mostly derived from iron removal thickener underflow (87–170 ppm U) and final neutralization thickener underflow (5–80 ppm U), according to waste monitoring data [9]. The activity concentrations of ^{238}U in the gypsum pond wastes range between 58 and 3375 Bq/kg [6], the latter exceeding the 1000 Bq/kg level above which, in terms of international radiation protection standards [10], the material would be regarded as radioactive for purposes of radiation protection. The main secondary minerals in the heaps are jarosite, goethite and gypsum. Goethite is uraniferous (730 ppm U on average), possibly resulting from the sorption of hexavalent uranium to goethite [6]. Uranium progeny (^{226}Ra, ^{210}Pb and ^{210}Po) mostly remain in the heaps during the leaching process, and are probably associated with secondary sulphate minerals, jarosite and gypsum [6]. High sulphate concentrations in the acidic PLS may limit the solubility of radium by incorporation in the crystal lattices of precipitated secondary sulphates. Thorium and progeny (^{232}Th, ^{228}Th, ^{228}Ra) are also mainly retained in the heaps [6].

2.2. Juomasuo and Hangaslampi gold–cobalt deposits

The Juomasuo and Hangaslampi gold-cobalt deposits are hosted by a metamorphosed volcanic sedimentary sequence in the Kuusamo schist belt (KSB), eastern Finland. Juomasuo is the largest known gold deposit in the KSB, averaging 4.6 ppm Au [11] and 158 ppm U [12]. The Hangaslampi deposit is located 900 m southeast of Juomasuo. Uraninite is the main uranium mineral in the Juomasuo and Hangaslampi deposits. Uraninite is found together with gold in fracture fillings and shear seams, and is erratically distributed throughout the gold lodes and sometimes within the cobalt mineralized zone. Uranium resources of the Juomasuo deposit, calculated on the basis of the data from the environmental impact assessment report [12], are 210 t uranium in the measured and indicated mineral resources of 1.5 Mt. Elevated uranium content (194–347 ppm U) is also found at Hangaslampi [12].

In recent years, Dragon Mining Ltd has investigated the possibility of developing a gold mining operation with a central processing facility and gold production from the deposits Juomasuo, Hangaslampi, Pohjasvaara, Meurastuksenaho, and Sivakkaharju. The uranium content is variable, and there are currently no plans for recovering uranium as a by-product. The planned gold production process includes mining, crushing, grinding, classification, flotation, cyanide leaching of gold, and tailings management. Crushed and ground ore is pumped to the flotation circuit to produce an intermediate gold concentrate. The fine grained tailings material (slurry), left after flotation, is pumped to the tailings management facility. Most of the uranium (65–80%) ends up in the tailings during flotation, the rest (20–35%) ending up in the flotation product [12]. The tailings material is divided via secondary flotation into two separate ponds depending on the sulphur content of the material (high and low sulphur tailings). It is also possible that uranium rich tailings will be separated into a third fraction via gravity separation. If uranium rich tailings were to be separated, about 10–15% of the uranium in the ore would remain in the low sulphur tailings, 20–35% in the high sulphur tailings, and 55–65 % in the uranium rich tailings [12].

2.3. Rompas gold prospect

Mawson Resources Ltd is focused on the Rompas exploration project in the Peräpohja Schist Belt in northern Finland, located a few kilometres south of the Arctic Circle in the municipality of Ylitornio. High grade gold and uranium have been found in the vicinity of Rompas-Rajapalot, over an area approaching 10×10 km. Rompas was discovered by Areva Resources Finland Oy in 2008 during a regional uranium exploration programme. Mawson acquired the project from Areva in 2010 and outlined the Rompas-type hydrothermal gold vein discovery over a 6 km strike length, with a width of 200–250 m. At Rompas, hydrothermal vein-type gold and uranium mineralization is mostly hosted by carbonate and calc-silicate veins in mafic metavolcanic rocks. The occurrences of gold minerals are confined to small pockets, having up to several thousand grams per tonne Au and up to tens of wt% U [13]. Gold is intimately associated with uraninite, typically in microfractures of uraninite.

2.4. Katajakangas Nb–REE deposit

The Katajakangas rare metal (Nb, REE, Zr, Ta) deposit, located in the Otanmäki area, central Finland, is a structurally controlled hydrothermal mineralization with fergusonite-(Y), ferrocolumbite, allanite and zircon as the main ore minerals. The deposit represents one of the potentially economic resources of Nb, REE, Zr and Ta in Finland. The occurrences of ore minerals are confined to narrow, fine grained, silicified zones 0.2–1.4 m thick, hosted by Paleoproterozoic gneissic granite. These mineralized zones have several hundred parts per million U and Th. In silicified zones, Nb and Ta are hosted primarily by ferrocolumbite and fergusonite-(Y), HREE by fergusonite-(Y), LREE by allanite, yttrium by fergusonite-(Y), and Zr by zircon. The deposit contains 0.76 wt% Nb_2O_5, 0.31 wt% Y_2O_3, 1.13 wt% ZrO_2 and 2.4 wt% Ln_2O_3 (lanthanides, i.e. rare earth elements excluding scandium and yttrium). Fergusonite is the dominant host to uranium, and allanite to thorium. The Katajakangas deposit has never been developed for mining, but an exploration licence covering the Katajakangas area is currently held by the private company Otanmäki Mine Oy.

2.5. Sokli phosphate deposit

Yara has recently planned to undertake phosphate mining in Sokli, eastern Lapland. The Sokli phosphate deposit is hosted by the Devonian Sokli carbonatite complex (365 Ma), which represents the westernmost intrusion of the Kola alkaline province. The Sokli ore is hosted by the surface weathered zone (regolith) of the carbonatite, with the ore mainly consisting of apatite. The average ^{238}U activity concentration of the Sokli phosphate ores is 310 Bq/kg, ranging from 100 Bq/kg to 1000 Bq/kg, whereas the average ^{232}Th activity concentration is 533 Bq/kg, ranging from 200 Bq/kg to 1700 Bq/kg [14]. Most uranium is bound to pyrochlore group minerals in the regolith, and thorium is mostly incorporated in pyrochlore group and monazite group minerals. There are currently no plans for recovering uranium as a by-product. During regolith formation, uranium was partially dissolved from pyrochlore due to weathering processes, and then fixed to some degree in secondary apatite within the regolithic phosphate ore [15]. In addition to the association with secondary apatite, uranium is also present to a lesser degree in fine grained goethite and altered residual magnetite within the regolith [16].

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RADIOLOGICAL ASSESSMENT OF TANTALITE MINING IN ETHIOPIA

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Abstract

A tantalite mine at Kenticha in the south of Ethiopia was visited for measurement of ambient radiation doses in the area and at workplaces in the mine and ore processing facilities. Radiation dose rates ranged from background values of 0.04 µSv/h, rising to 0.08 µSv/h in the mine excavations and up to 0.12 µSv/h near the minerals sieving and hydraulic sorting tables. Higher dose rates were measured near the tantalite concentrate drying work station (0.20 µSv/h) and in the tantalite storage warehouse, reaching 22.5 µSv/h near the tantalite concentrate piles. The elevated dose rates were due to uranium and thorium radionuclides co-occurring in the tantalite ore. Analyses of samples performed in the laboratory indicated that in the dry ore concentrate the ^{238}U activity concentration was 53 810 ± 4570 Bq/kg and the ^{232}Th concentration was 515 ± 73 Bq/kg. The operation of weighing and packaging tantalite concentrate in drums and the loading of drums onto a truck was monitored and radiation doses received by mine workers and transport workers were assessed. Occupational and environmental radiation exposure levels are discussed.

1. INTRODUCTION

Tantalite is a mineral of tantalum, a corrosion resistant non-radioactive metal widely used in microelectronics. Production of tantalite has been increasing worldwide and new mines are expected to start production in several regions. Ethiopia is a tantalite ore producer, mainly based on the production of Kenticha mine in the south of the country which produces about 300 t/a of tantalum concentrates consisting of colombite–tantalite and other Ta oxides. The mine is situated in the Kenticha pegmatite field in which granitic pegmatites cluster over an area of roughly 2500 km^2. Most of the intruding pegmatite bodies range in size from a few metres to more than 1 km in length. The pegmatites have been classified into rare-earth-element-enriched and barren feldspar–muscovite and have been dated to around 530 million years, which connects them to regional post-orogenic granite magmatism. As the mineral reserves in Kenticha have been estimated at 20 000 t of Ta_2O_5 and Nb_2O_5, mining in this area is set to expand [1]. Little information has been published in international scientific literature about the co-occurrence of natural radioelements in tantalite ores, although tantalum minerals have been identified in IAEA reports as NORM [2, 3]. In 2015 a radiological survey and sampling was carried at the Kenticha mine site. The results of field measurements and analyses of radionuclides are presented.

2. MATERIALS AND METHODS

Kenticha mine is located in a remote mountainous area in the south of Ethiopia. Opencast mining operations are carried out on the mountainside by the Ethiopia Mineral Development Enterprise. The overburden is removed and deposited nearby, while the tantalite rich layer is excavated (see Fig. 1) and transported by truck to the mineral feeder for sieving and stone removal, followed by sorting of the heavy minerals on hydraulic shaking tables. The segregated denser minerals, including iron and other magnetic metals, are further separated by magnetic drums, leaving a wet concentrate of tantalite which is dried on trays over gas burners and then transported to a storage warehouse on the mine premises. The remaining minerals are stored as a slurry in a dam on site, pending further processing to extract residual tantalite. Materials rejected from this further processing are disposed of into another dam.

FIG. 1. Tantalite mining at Kenticha mine (left); weighing and packaging of tantalite concentrate for transport by road (right).

In preparation for the transport of the dry tantalite concentrate by road, the material is packaged into drums. The drums are weighed, carried on shoulders to the yard outside and loaded onto a truck. This packaging, weighing and loading operation occupies about 10 workers for several hours.

The entire mine concession area was monitored for gamma radiation. Dose rates were measured with a portable radiation monitor FH40 (Thermo Scientific) and a portable gamma spectrometer Identifinder (FLIR). Samples of solid materials were collected at selected points, transferred to plastic bags and taken to the laboratory for radionuclide analysis by radiochemical separation procedures followed by alpha spectrometry of radionuclides electroplated on stainless steel discs [4].

3. RESULTS AND DISCUSSION

The results of the average ambient dose rate measurements are shown in Table 1. Exposures to tantalite ore at the work stations in the hydraulic sorting area were sufficiently low as to be unlikely to give rise to annual doses exceeding the 1 mSv dose limit for non-radiation workers. Exposures to tantalite concentrate during packaging, loading and transport were higher and may give rise to doses exceeding this value.

The results of the analysis of the various materials associated with the operation are shown in Table 2. Soils collected near the mine area and geologic materials collected in the mine excavation area indicated the presence of uranium and thorium in similar concentrations in bulk minerals and in soils. The ^{238}U concentrations in the soils in the region were found to be 111 ± 3 Bq/kg. Similar activity concentrations were found for ^{238}U decay progeny. After sorting and concentrating the tantalite ore, uranium was found to be associated with the tantalum containing fraction at concentrations higher than those of thorium. Gravel remaining in the mine workings after excavation displayed activity concentrations lower than those in unmodified topsoils — the uranium content was lower and the ^{238}U activity concentration was 69 ± 2 Bq/kg. The iron fraction, after magnetic separation from raw tantalite ore, was still rich in uranium, with a ^{238}U activity conentration of 254 ± 8 Bq/kg. After further processing of materials remaining from the initial separation of tantalite concentrate, the waste material disposed of into the waste pond, although containing little tantalum, was found to be richer in uranium than expected. The activity concentration of ^{238}U in the tantalite concentrate was very high, at 53 810 ± 4570 Bq/kg, while the activity conentration of ^{232}Th was 15 ± 73 Bq/kg, indicating a much higher association of uranium relative to thorium in the tantalite concentrate.

TABLE 1. DOSE RATES AT VARIOUS LOCATIONS

	Dose rate (μSv/h)
Outside the mine concession area	0.040 ± 0.005
On the mine premises near the canteen and administration buildings	0.06 ± 0.01
Mine excavations	0.10 ± 0.02
Mineral sorting tables	0.10–0.12
Iron magnetic separator and tantalite drying pans	0.12–0.20
Above the pond for storage of rejected minerals prior to further processing	0.20
Above the pond for disposal of process waste	0.07
Outside locked warehouse for storage of dried tantalite concentrate	1.43 ± 0.03
Ore pile	22.5
Surface of drums containing dry tantalite concentrate	14
Next to truck loaded with drums of dry tantalite concentrate	Up to 8.2
Driver’s seat of truck loaded with drums of dry tantalite concentrate	1.7

TABLE 2. RADIONUCLIDE CONCENTRATIONS IN VARIOUS MATERIAL SAMPLES

	Mean activity concentration and standard deviation (Bq/kg)			
	^{238}U	^{230}Th	^{226}Ra	^{232}Th
Soil from above mine area	111 ± 3	85 ± 5	94 ± 10	9.7 ± 0.9
Material in new excavation	106 ± 4	111 ± 7	202 ± 16	46 ± 3
Material in old excavation (gravel)	69 ± 2	41 ± 8	71 ± 4	17 ± 5
Iron fraction	254 ± 8	187 ± 13	202 ± 30	30 ± 3
Tailings in storage dam pending further processing	274 ± 8	188 ± 10	137 ± 8	88 ± 5
Tailings in disposal dam after further processing	142 ± 4	104 ± 5	104 ± 5	40 ± 2
Tantalite concentrate	53 810 ± 4570	36 271 ± 5018	29 416 ± 4528	515 ± 73

4. CONCLUSIONS

The tantalite ore contains uranium and thorium in concentrations that initially are low in the geological deposit and mine products. After ore processing, most of the radioactivity ends up in the tantalite concentrate at high activity concentrations. The operations carried out in the mining and ore sorting phases do not seem to generate high radiation doses at workplaces, including those at the excavation pit and at the sieving and hydraulic mineral sorting facilities. However, radiation doses are higher at the ore drying station and really become of radiological concern near the tantalite concentrate piles stored in the warehouse.

During the operations of ore packaging, loading and transport by road, the exposure of workers may become significant and is likely to exceed the dose limit for non-radiation workers as specified in the International Basic Safety Standards [5]. Therefore, radiation protection procedures should be included in the routine tasks of tantalite mining at Kenticha. Furthermore, as all workers live in the mine concession area and grow vegetables and graze animals in the neighbourhood for family consumption, an in-depth radiological risk assessment, including an assessment of the exposure of members of the public through ingestion of food and water, should be carried out.

Mineral extraction, and in particular the development of the Kenticha tantalite mining project, is of high economic value for the country. The Ethiopian Radiation Protection Authority is taking steps to ensure adequate radiation protection of workers and members of the public.

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RADIOLOGICAL INVESTIGATION AT YATAGAN COAL FIRED POWER PLANT, TURKEY

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Abstract

Coal has a significant role in energy production worldwide. It contains radionuclides of natural origin such as ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K and trace elements such as Cd, Cr, Pb, Ni and Zn. These radionuclides and trace elements become enriched in ash fractions during the combustion process in coal fired thermal power plants. Therefore, these plants can be major sources of increased natural radioactivity in the environment. In this study, coal and its combustion residues (flyash and bottom ash) generated in the Yatagan coal fired thermal power plant in Mugla, Turkey were analysed by gamma-ray spectrometry to determine the activity concentrations of radionuclides of natural origin. Samples of flyash, a material widely used as a cement additive for construction purposes, were collected at different stages along the emission control system of the thermal power plant. Calculation of the 'activity index' for each sample gave an indication of the risk due to external gamma exposure that would result from the use of the material as a component of construction materials.

1. INTRODUCTION

Radionuclides of natural origin are released into the environment from various anthropogenic sources such as the phosphate industry, oil and gas production, metal mining and smelting, combustion of fossil fuels and other energy production [1–6]. Such industries often produce large amounts of NORM residues. Some of these residues, for instance coal ash and phosphogypsum, may be used in building materials such as cement, concrete, aerated concrete, bricks and lightweight aggregate, and in agriculture [7–9].

The residues produced by the combustion of fossil fuels such as coal and lignite contain radionuclides of natural origin (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K) and the by-product use of such residues may lead to a significant increase in human exposure. The radiological risk depends on various factors, such as the concentrations in the coal, the ash content, the combustion temperature and radionuclide partitioning between bottom ash and flyash. In recent years, there have been many studies on the utilization of coal flyash as an industrial by-product in order to prevent environmental pollution caused by this residue. The storage of coal combustion residues from thermal power plants leads to significant environmental problems. Therefore, the use of these residues will have economic and resource conservation advantages.

However, coal fired thermal power plants that produce NORM during their operation should be investigated from the radiological point of view as an additional radiation source. Before using these residues it is important to evaluate their physical parameters, chemical composition and radiological characteristics. The radionuclides from the ^{238}U series have a different behaviour and enrichment at different stages of the combustion process [5, 6, 10]. The aim of this study was to determine the concentrations of radionuclides of natural origin (^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{230}Th, ^{232}Th and ^{40}K) in coal and ash samples collected at different stages of the emission control system at the Yatagan thermal power plant and to assess the implications for using flyash in particular as a component of building material.

2. MATERIALS AND METHODS

2.1. Sampling

This study focuses on the Yatagan coal fired thermal power plant located in the south-west of Turkey. The plant was established in the district of Yatagan in Mugla province, which has 415 255 000 t of lignite reserves. The average daily coal requirement of the plant is 18 000 t. The chimney height of the plant is 120 m. Yatagan thermal power plant has three units producing 210 MW each. The units started working between 1982 and 1984. South Aegean Lignite mine, which is a subsidiary of the General Directorate of Turkish Coal Enterprises (TKI), is responsible for obtaining the coal from mines to use at the Yatagan thermal power plant. According to the data provided by the Turkish State Meteorological Service, prevailing wind directions are south-eastern, north-eastern and south-western.

Coal, bottom ash and flyash samples were collected from the Yatagan thermal power plant. The flyash samples were collected at the water pre-heater (economizer), air pre heater (LUVO) and electrostatic precipitators (ESP) along the emission control system, which is shown in Fig. 1. All flyash fractions are stored temporarily in stockpiles (SILO). Flyash samples were also collected at the SILO point.

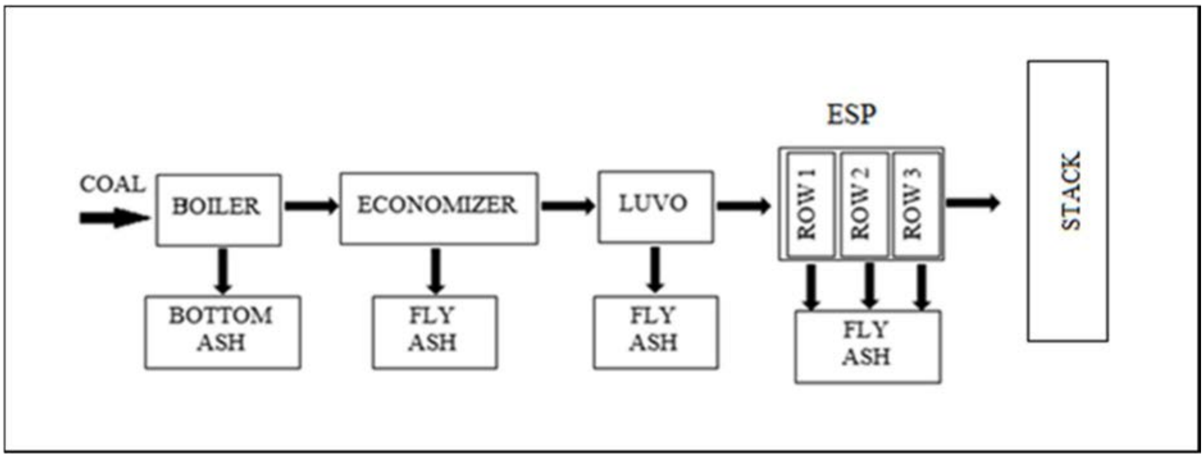

FIG. 1. Sampling points within the power plant.

2.2. Gamma spectrometry

All samples were measured by gamma spectrometric techniques for the determination of ^{238}U (^{234}Th), ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{230}Th, ^{232}Th and ^{40}K. The samples were placed into aluminium cases and dried at 105°C for 24 h. The dried samples were compacted by a hydraulic press and placed into metal containers with a volume of 55 cm^3 for gamma spectrometric analysis. To achieve secular equilibrium in the ^{238}U series (between ^{226}Ra and its progeny) and in the ^{232}Th series (between ^{228}Th and its progeny), the samples were sealed hermetically to avoid ^{222}Rn and ^{220}Rn emanation. The sealed samples were stored for at least 23 days (6–7 half-lives of ^{222}Rn) before making the gamma spectrometric measurements. A high-purity germanium detector (Canberra BeGe BE 3830-P) was used to measure the activities of selected radionuclides. The measurement energy calibration was done using an RGU-1 source. Three sources (RGU-1, RGTh-1 and RGK-1, which are IAEA certified reference materials) were used in efficiency calculations. The activity concentration of ^{210}Pb was determined according to its 46.5 keV gamma energy peak. A self-absorption correction was performed for determining ^{210}Pb and ^{234}Th activity concentrations according to Ref. [11].

3. RESULTS AND DISCUSSION

The results of the analysis of coal and ash fractions are shown in Fig. 2. The ranges of activity concentrations were as follows:

^{238}U: 155 ± 12 to 451 ± 20 Bq/kg;
^{226}Ra: 187 ± 7 to 563 ± 20 Bq/kg;
^{228}Ra: 67 ± 4 to 159 ± 7 Bq/kg;
^{210}Pb: 186 ± 20 to 1153 ± 44 Bq/kg;
^{230}Th: 162 ± 60 to 531 ± 88 Bq/kg;
^{232}Th: 65 ± 3 to 136 ± 5 Bq/kg;
^{40}K: 305 ± 18 to 831 ± 29 Bq/kg.

Our sampling method provides a clear understanding of the process of enrichment of radionuclides. Since ^{210}Pb is generally more volatile than other natural radionuclides, it is generally more enhanced in the flyash samples in the electrostatic precipitators of the plant. The activity concentrations of ^{226}Ra and ^{210}Pb in flyash collected along the emission control system of the plant have different levels of enrichment. While the concentration ratio of ^{210}Pb to ^{226}Ra was found to be 0.40 at the water pre-heater point (economizer), the ratio was 2.05 at SILO where all flyash fractions were combined. The minimum concentration ratio of ^{238}U to ^{226}Ra was found to be 0.65 at SILO. This ratio indicates that there are significant differences in radionuclide enrichment inside the power plant. It may be linked to the different particle sizes of fly ash and the combustion conditions.

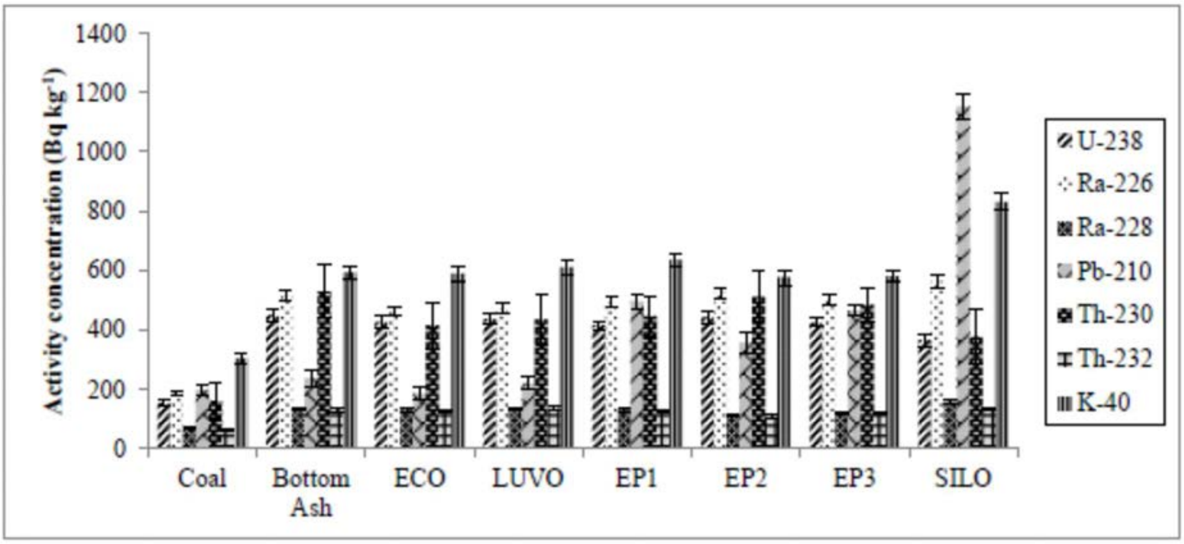

FIG .2. Radioactivity in ash fractions.

The use of flyash as a by-product for use in building materials is a source of gamma exposure owing to its radioactivity content. Concrete is considered as the main building material in most countries. It is made from raw materials that contain radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K. The activity concentrations of these radionuclides found in cement can be significantly lower in comparison with those in flyash and some other industrial by-products. Nowadays, there are many studies on the use of flyash produced from power plants in concrete mixes as a partial replacement for cement. An activity index (*I*) is referred to in the European Directive 2013/59/Euratom as a means for controlling radiation exposures in building material containing NORM, such as flyash. The index is based on the activity concentrations of the three radionuclides ^{232}Th, ^{226}Ra (from the ^{238}U decay series) and ^{40}K, as follows:

$$I = \frac{C_{\mathrm{Th}}}{200} + \frac{C_{\mathrm{Ra}}}{300} + \frac{C_{\mathrm{K}}}{3000} \tag{1}$$

where C_{Th}, C_{Ra} and C_{K} are the concentrations in becquerels per kilogram of ^{232}Th, ^{226}Ra and ^{40}K, respectively. If the activity index does not exceed 1, the materials can be used without further radiological assessments. In this study, the activity index of the flyash was calculated using the above equation in all samples which were taken at the boiler, water pre-heater (economizer), air pre heater (LUVO) and electrostatic precipitators (ESP). The lowest activity index was exhibited by the sample taken from the water pre-heater (ECO) point — assuming 30% use of the flyash in cement, the activity index of the mixture was found to be 0.71. For flyash sample taken from the SILO point, the activity index was calculated to be 0.85.

4. CONCLUSIONS

This work provided new data on NORM activity concentrations in ash collected at different stages along the emission control system at Yatagan coal fired thermal power plant in Mugla, Turkey. The use of flyash in construction materials is a well known source of gamma exposure due to the presence of radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K. According to the results obtained in this study, if the building materials were to be prepared from the ash at a proportion not exceeding 40%, any radiological issues in terms of external gamma radiation can be ignored.

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Poster Presentation

RADIOACTIVITY IN SOIL NEAR OILFIELDS

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Abstract

The concentrations of radionuclides of natural origin in soil samples from Kuwait and Qatar were measured using gamma spectroscopy and the results were used to calculate several other parameters related to radiological risk. It was found that most samples were within the normal range for rocks and soil worldwide, but samples from locations near oilfields were found to have enhanced values due to past oil extraction activity in the area.

1. INTRODUCTION

Radionuclides of natural origin are divided into two main groups: cosmogenic radionuclides such as ^{14}C, ^{7}Be and ^{26}Al, and primordial radionuclides (groups of radionuclides forming the ^{238}U, ^{235}U and ^{232}Th decay chains and single radionuclides such as ^{40}K) that are present in the Earth's crust [1]. For each of the terrestrial decay series radionuclides, some of the decay products emit characteristic gamma rays. These emissions do not in themselves represent gamma decay but are rather a mechanism for emitting excess energy associated with other modes of decay. The measurement of these gamma rays using gamma spectroscopy allows us to determine the activity concentrations of each series and to identify the specific radionuclides which decay [2]. The purpose of this study was to determine the activity concentrations in several soil samples from Kuwait and Qatar and to establish some idea of the levels of radiological hazard associated with the radioactivity in the samples.

2. METHOD

Soil samples were collected from Kuwait (nine samples) and Qatar (three samples). The procedures for preparing the soil samples to make them homogenous and to remove the impurities involved drying, particle size control and storage. To dry the soil at room temperature, each sample was kept in at oven at 50°C for 24 h. To control the particle size, each sample was passed through a sieve to remove oversize impurities. Each sample was then placed in a 550 mL Marinelli beaker and sealed and stored for 4 weeks to enable equilibrium conditions to be attained. Background measurement was taken by using deionized water in a Marinelli beaker. The sources, deionized water and soil samples had the same geometry to reduce errors in calculation due to gamma-ray attenuation. The Marinelli beakers, deionized water and soil samples are shown in Fig. 1.

A high purity germanium detector used for gamma spectrometric analysis was characterized using four standard sources to calibrate the energy and efficiency of the detector. The sources were an NG3 mixed source, ^{152}Eu, ^{232}Th and ^{226}Ra. These sources have a wide range of energy from 59.9 to 2614 keV. This range is important for the detection of the radionuclides in the samples. Each sample was measured for 48 h to collect the radionuclide counts using gamma spectroscopy. For the ^{238}U and ^{232}Th series, the radionuclides and the energy peaks measured were as follows:

Uranium-238 series: ^{226}Ra (186.2 keV), ^{214}Pb (242, 295 and 351 keV) and ^{214}Bi (609.3, 1120.2, 1238.1, 1764.4 and 2204.2 keV);

Thorium-232 series ^{212}Pb (238.6 keV), ^{228}Ac (270.3, 338.3, 911.2 and 968.9 keV) and ^{208}Tl (583.1 and 2614.7 keV).

FIG. 1. Marinelli beakers, deionized water and soil samples

3. RESULTS

The results of the activity concentration measurements are shown in Table 1. For all samples, the weighted means were calculated for the ^{238}U and ^{232}Th series (see for example Figs 2 and 3) and for the single radionuclides ^{40}K and ^{137}Cs.

TABLE 1. RADIOACTIVITY IN SOIL SAMPLES

Sample code	Activity concentration (Bq/kg)			
	^{238}U series	^{232}Th series	^{40}K	^{137}Cs
064-F12	16.89 ± 0.18	6.36 ± 0.19	92.8 ± 3.1	4.08 ± 0.11
076-G11	15.65± 0.16	13.84 ± 0.13	210.5 ± 6.6	0.52 ± 0.09
173-Q01	6.22 ± 0.11	6.66 ± 0.11	236.5 ± 7.4	1.83 ± 0.07
23-X-288	50.69 ± 0.51	3.2 ± 0.03	80.5 ± 2.5	0.98 ± 0.44
29-X-228	339.31 ± 3.54	8.83 ± 0.11	162.7 ± 5.1	1.69 ± 0.75
2-X-228	13.63 ± 0.14	5.65 ± 0.06	169.8 ± 5.3	0.17 ± 0.074

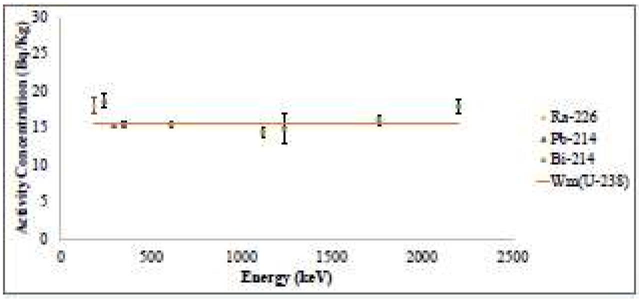

FIG. 2. Determination of weighted mean activity concentration for ^{238}U series radionuclides in Sample 076-G11.

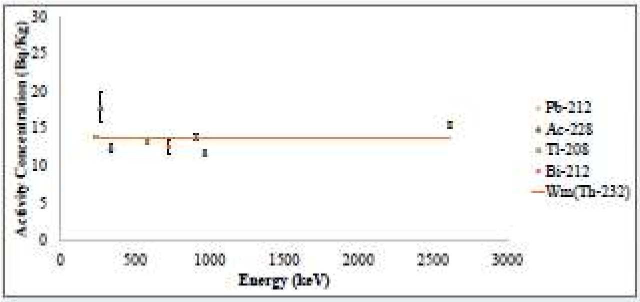

FIG. 3. Determination of weighted mean activity concentration for ^{232}Th series radionuclides in Sample 076-G11.

The following radiological parameters for the samples were calculated: radium activity equivalent (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), absorbed dose rate and annual effective dose. The results are shown in Table 2. The calculated annual effective dose for all samples was below 1 mSv. The samples which had been collected in areas close to oilfields had different enhanced values as a result of industrial processes in these regions such as disposal of scales and sludge in pipes into the surrounding environment.

TABLE 2. RADIOLOGICAL PARAMETERS FOR THE SOIL SAMPLES

Sample code	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	Absorbed dose rate (nGy/h)	Annual effective dose (µSv/a)
064-F12	33.1 ± 2.1	0.089 ± 0.006	0.57 ± 0.04	15.5 ± 0.9	19.1 ± 1.3
076-G11	51.6 ± 3.2	0.139 ± 0.007	1.23 ± 0.09	24.3 ± 1.9	29.9 ± 1.7
173-Q01	33.7 ± 2.2	0.091 ± 0.007	1.32 ± 0.10	16.6 ± 1.1	20.4 ± 1.5
23-X-288	61.4 ± 4.8	0.166 ± 0.013	0.58 ± 0.04	28.7 ± 2.3	35.2 ± 2.9
29-X-228	364.7 ± 27.4	0.986 ± 0.072	1.83 ± 0.17	169 ± 15	207 ± 17
2-X-228	34.8 ± 2.3	0.094 ± 0.006	0.97 ± 0.08	16.8 ± 1.1	20.6 ± 1
Range of worldwide mean values	≤370	≤1	≤1	18–93	

4. DISCUSSION

The activity concentrations in the samples from Qatar were higher than those in the samples from Kuwait. The samples from Dukhan region, located in the north-west of Qatar, had the highest ^{226}Ra values. This area has an old inshore oil well. As expected, the ^{40}K concentrations, ranging from 92.9 ± 3.1 to 258 ± 8 Bq/kg, reflected the natural abundance of potassium (and thus ^{40}K). The anthropogenic radionuclide ^{137}Cs which comes from nuclear reactors, nuclear weapons tests (fallout) and nuclear accidents was found in very low levels in all samples.

The radiological parameters calculated for all samples from Kuwait and Qatar fell within the worldwide range of normal rocks and soil except for one sample 29-X-228 which was significantly above this range [3].

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Poster Presentation

EVALUATION OF NORM RESIDUES FROM THE PROCESSING OF CRUDE OIL IN A REFINERY

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Abstract

In the processing of crude oil in a refinery, over 1000 t of NORM residues per year can result. For the use or disposal of these residues, normally the control limits must not be exceeded. For the evaluation of accumulated residues, different samples were taken in an oil refinery. The highest ^{210}Pb activities were measured on scaling of heat exchangers (about 8000 Bq/kg) and installed equipment in distillation columns (about 25 000 Bq/kg). In Germany the 95 % upper confidence limit of the mean of a random sample specific activity has to fall below 1000 Bq/kg concerning dumping or combustion of NORM residues. Several samples were taken from sewage sludge (800 t/a) and oil sludge (500 t/a) as independent random samples. From these samples the upper confidence limit for the confidence level 0.95 has been determined by means of classical and modern numerical ('bootstrapping') statistical methods. In the case of ten independently removed samples of oil sludge, the control limit was exceeded in terms of both statistical methods – in the classical and the numerical as well. When the sample size was expanded to 20 samples, by means of the bootstrap method it could be shown that the control limit was not exceeded. By using the classical method, the control limit was exceeded again as previously.

1. INTRODUCTION

In oil refineries, incoming crude oil has normally a low activity (<40 Bq/kg per radionuclide). During processing, however, individual radionuclides are enriched. As a result of the activity flow several radionuclides can become concentrated in the residues. Over 1000 t of NORM residues per year accumulate. For the reuse (road construction, house construction etc.) or disposal (dump, combustion) of these residues, normally the control limits must not be exceeded. For the evaluation of these accumulated residues, different samples were taken in the refinery (e.g. oil sludge, sewage sludge, used catalyst, coke, bitumen, sulphur, furnace linings).

2. METHODS

In the case of landfill disposal or combustion of NORM residues in Germany, the dose received by members of the public must not exceed 1 mSv/a. This level is automatically kept if the residues meet the following condition:

$$C_{U\text{-}238max} + C_{Th\text{-}232max} \leq 1000 \text{ Bq/kg}$$

$C_{U\text{-}238max}$ and $C_{Th\text{-}232max}$ are the largest concentrations of activity of all radionuclides within the respective decay chain. If $C_{U\text{-}238max}$ or $C_{Th\text{-}232max}$ is <200 Bq/kg the corresponding chain remains unconsidered. If random samples are selected, the following applies:

$$C_{U\text{-}238max} \equiv {}^{95\%}UCL(C_{U\text{-}238max}); \qquad C_{Th\text{-}232max} \equiv {}^{95\%}UCL(C_{Th\text{-}232max});$$

where the representative value $^{95\%}$UCL is the upper confidence limit (UCL) of the estimation of expected value (or mean) regarding the activity concentration for the confidence level 0.95 ($^{95\%}$UCL). The expected value of a population characterizes the mean of this population. The evaluation of the random sample is carried out by comparison of the test statistic $^{95\%}$UCL with the control limit (in this case 1000 Bq/kg). Classical and modern numerical statistical methods can be used to determine the confidence limits for the mean of normal or lognormal distributions. In addition to classical statistics, we use ‘bootstrapping’. Regarding methodology and formulas in this matter we refer to Refs [1–3].

3. RESULTS

For the evaluation of accumulated residues and for the investigation of activity flow within the refinery, 49 samples of residues, plant components and media were taken (see Fig. 1). Some of the gamma-spectrometric analysis results are shown in Table 1. The results show a correlation between the measured activities and the prevailing operating temperatures (desalter: 130–250°C; fluid catalytic cracking (FCC): 500–760°C; heat exchanger: 400°C).

Lead-210 proved to be the dominant radionuclide. The highest ^{210}Pb activities were measured on scaling of heat exchangers (8000 Bq/kg) and installed equipment in distillation columns (25 000 Bq/g). From the final residues, the sewage sludge (800 t/a) and oil sludge (500 t/a) proved to be relevant regarding the quantity. The ^{210}Pb activity concentrations of ten samples from sewage sludge did not exceed 60 Bq/kg and one sample had 285 Bq/kg. Therefore sewage sludge is clearly under the control limit of 1000 Bq/kg. From the oil sludges ten samples were taken at first. When the ^{210}Pb activity is higher than a factor of five above the highest activity of the remaining radionuclides in the decay chain, then you can halve ^{210}Pb and the following applies: $C_{\text{Pb-210eff}}$: = 0.5 × $C_{\text{Pb-210}}$ (see Table 2). Since the limit was exceeded ($^{95\%}$UCL = 1154 Bq/kg for $\text{Pb-210}_{\text{eff}}$), an additional ten samples were taken (see the values in Table 2 and the histogram in Fig. 2). According to the quantile–quantile (Q–Q) plot, a lognormal distribution is likely (R^2 = 0.93, see Fig. 3).

FIG. 1. Residues, components and media of the refinery.

TABLE 1. RADIOACTIVITY IN REFINERY RESIDUES, COMPONENTS AND MEDIA

Radio-nuclide	Activity concentration (Bq/kg)										
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Sample 7	Sample 8	Sample 9	Sample 10	Sample 11
Th-232 series:											
Th-232	–	–	6	–	–	31	–	–	–	–	–
Ra-228	29	23	164	0.03	109	131	40	275	16	–	–
Th-228	18	25	158	0.02	99	90	38	162	16	5	4
U-238 series:											
U-238	11	16	7	0.3	52	30	24	83	12	21	–
Th-230	–	17	31	–	–	–	30	103	–	–	–
Ra-226	70	62	455	0.3	170	175	48	391	22	–	14
Pb-210	285	1039	3130	1	444	1961	63	8411	170	259	24 567
U-235 series:											
U-235	0.6	0.9	0.4	0.07	2.2	1.9	1.1	3.7	0.7	–	–
Ac-227	0.3	0.8	0.6	0.04	6.0	3.8	1.0	0.2	2.4	–	–
Pa-231	0.3	–	–	–	6.1	3.5	2.3	–	0.6	–	–
K-40	10	34	37	0.1	48	36	58	39	13	–	–

Note: Samples were as follows: 1: Sewage sludge (conditioned); 2: Oil sludge; 3: Desalter sludge; 4: Sewage water; 5: FCC catalyst withdrawal (E-cat); 6: FCC catalyst (dust out of electrostatic precipitator); 7: Bitumen; 8: Oil sludge deposition on heat exchanger bundle; 9: Coke; 10: Distillation column internals (deposit on trays); 11: Distillation column internals (deposit on packings).

TABLE 2. LEAD-210 CONCENTRATIONS IN OIL SLUDGE

Sample	Activity conentration (Bq/kg)	
	Pb-210	Pb-210_{eff}
1	5	5
2	4288	2144
3	52	52
4	10	10
5	99	99
6	42	42
7	4776	2388
8	3	3
9	8	8
10	179	90
11	396	198
12	1039	520
13	282	141
14	183	92
15	163	163
16	359	180
17	372	186
18	278	139
19	110	110
20	632	316

Note: $C_{\text{Pb-210eff}} = R \times C_{\text{Pb-210}}$, where $R = 1$ ($C_{\text{Pb210}}/C_{\text{U-238max}} \leq 5$) or R=0.5 ($C_{\text{Pb210}}/C_{\text{U-238max}} > 5$).

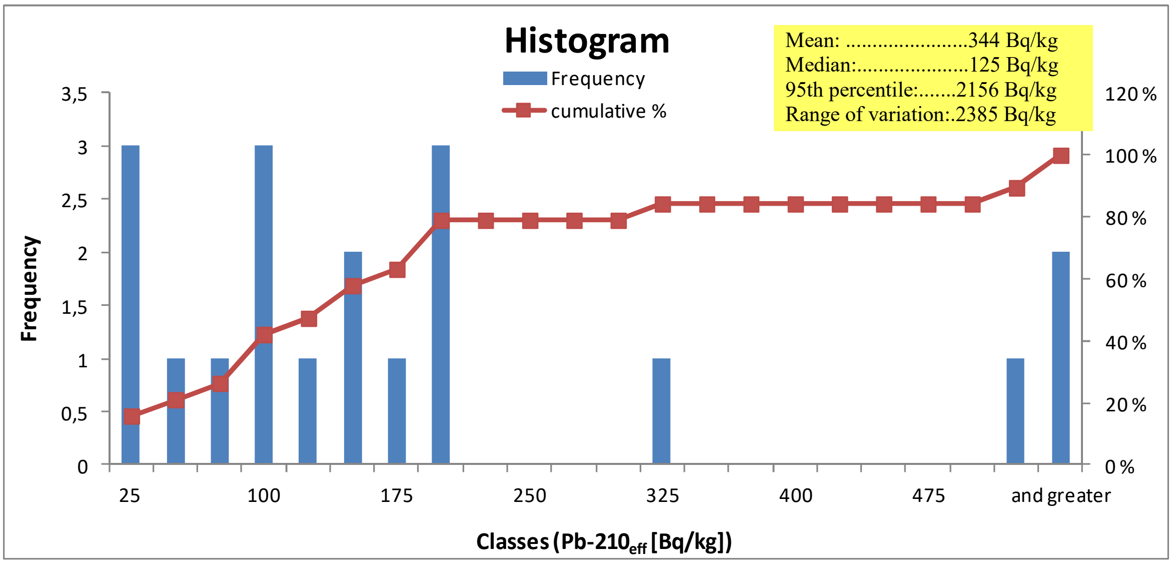

FIG. 2. Frequency distribution of $^{210}Pb_{eff}$ *for 20 random oil sludge samples.*

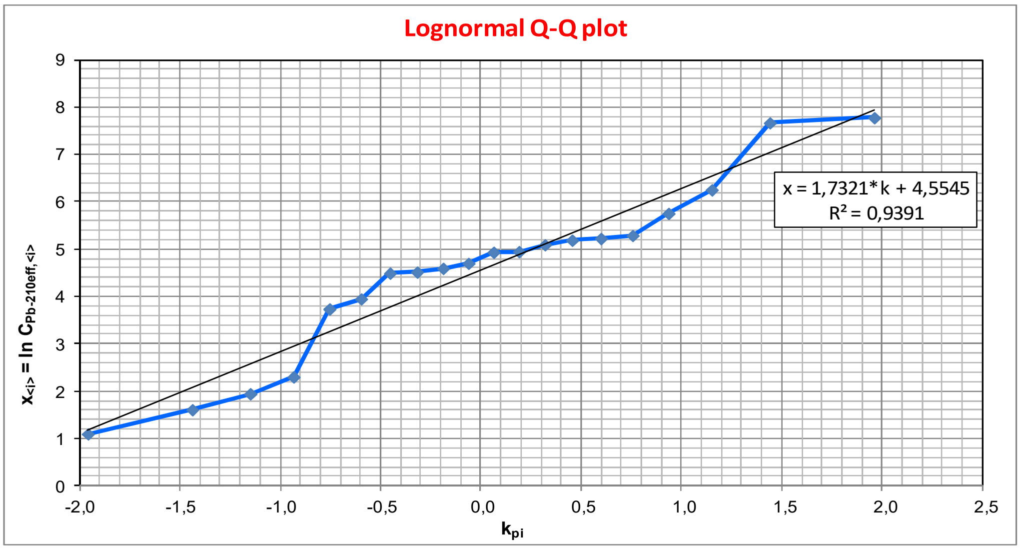

FIG. 3. Q–Q plot for the logarithm of the measured values regarding the mass-related activity of $^{210}Pb_{eff}$ *and calculation of the 'coefficient of determination'* $R^2_{(Ln)}$.

From the random sample of size *n* = 20 taken from a population of oil sludge samples, the $^{95\%}$UCL was determined for the dominant radionuclide ^{210}Pb$_{eff}$ at first conventionally and then by using the 'bootstrap' method with BCa (bias correction and acceleration) modification. Table 3 shows the results. By using the classical method, the control limit was exceeded again as previously. Although the arithmetic average is just 344 Bq/kg and the estimation for the expected value (maximum likelihood estimator) is just 461 Bq/kg, the $^{95\%}$UCL is 2142 Bq/kg. The latter has a relatively high confidence factor of 4.65 due to the highly expanded frequency distribution to the right (see Fig. 2). A $^{95\%}$UCL of 798 Bq/kg resulted by means of the bootstrap method. In this way the control limit was observed.

TABLE 3. ANALYSIS OF ^{210}Pb$_{eff}$ FOR 10 AND 20 RANDOM SAMPLES OF OIL

SLUDGE

	Pb-210$_{eff}$ (Bq/kg)	
	10 samples	20 samples
Arithmetic mean	484 Bq/kg	344 Bq/kg
$^{95\%}$UCL (classical)	No realistic value, size n = 10 too small	2142 Bq/kg
$^{95\%}$UCL (numerical)[a]	1154 Bq/kg	798 Bq/kg

[a] Bootstrap method with B = 1000 replications.

4. DISCUSSION

In Ref. [3] the bootstrap method was tested on the basis of examples. Realistic $^{95\%}$UCL values were generated, which were closer to the expected value than in the case of using the classical statistics. The method proved to be a suitable, consistent, efficient, robust and parameter-free method.

5. CONCLUSIONS

By means of random samples, a collection of measurement results is generated. To assess the measured values, the test statistic $^{95\%}$UCL is conservative enough. If the $^{95\%}$UCL exceeds the control limit, there are several ways to reduce it: increase of sample size, higher measurement accuracy, replacement of classical statistical approaches by modern numerical methods (e.g. bootstrapping).

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Poster Presentation

RADIOACTIVITY IN GAS MANTLES SOLD IN MEXICO

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Abstract

Thorium-containing incandescent mantles used for gas lighting are considered sources of significant radioactive exposure of the public and environment and are therefore recognized as NORM products. In countries with NORM regulations, radioactive gas mantles are subject to control to avoid undue health hazards to consumers or radiological contamination of the environment. In Mexico, the commercial sale of radioactive gas mantles is not regulated from a radiological point of view and these products are widely used for activities such as the illumination of street food stalls. In this work, the radiometric characterization of gas mantles sold in Mexico is presented. The methods used for characterization were high resolution gamma-ray spectrometry using a high purity germanium detector and alpha-particle spectrometry using a solvent extraction technique. The results indicate that high concentrations of natural radionuclides from the thorium decay series are present in some of the gas lantern mantles analysed, as well as in the waste generated, and may represent a radiological risk to exposed members of the public.

1. INTRODUCTION

Thoriated mantles for gas lamps have been used for interior and exterior lighting since the late 19th century. The manufacturing process involves the dipping of a highly absorbent rayon stocking into a solution containing mostly thorium nitrate. The thorium content is derived from minerals that generally contain not only thorium but some uranium as well. Some producers have now replaced thorium by non-radioactive elements such as yttrium and zirconium. Nevertheless, thoriated gas mantles are still legally marketed in several countries. They are considered NORM products because their thorium content could result in a significant increase in exposure of individuals. In European countries, the sale of thoriated gas mantles is prohibited or subject to licensing if the thorium content exceeds a certain value, but such products are not subject to regulation in Mexico and can therefore be sold without any restrictions. In addition to camping applications, people in Mexico make extensive use of gas lamps for the illumination of street food stalls at night. The labeling of thoriated gas mantles sold in Mexico provides no indication of the radioactivity content and mantles that have reached the end of their useful life are disposed of in normal household waste.

This paper describes the assessment of thoriated gas mantles used in Mexico in terms of their radiological impact on workers and consumers. The radiometric characterization of gas mantles was conducted by two different and complementary techniques: gamma-ray spectrometry and alpha-particle spectrometry.

2. METHOD

2.1. Samples

Several commercially available brands of thoriated gas mantle were purchased from various regions of Mexico. On the labels of some of the mantles, it was stated that the product was made from 100% silk and it was recommended that the product be disposed of in household waste at the end of its life. For comparison purposes, some gas mantles were purchased in Spain — the labelling on these products indicated clearly that they were not radioactive. The gas mantle samples were compressed and vacuum sealed in cylindrical polypropylene containers and stored for more than 30 d to make up for any radon depletion and to ensure secular equilibrium between ^{224}Ra and its short-lived progeny.

Three 300 mg samples of ash generated by gas mantles were also obtained and sealed in special vials. The first sample (A-MxFS) consisted of ash collected directly from street food markets in Mexico. The second sample (A-GMx6) consisted of ash obtained from the combustion in the laboratory of sample GMx6, a thoriated gas mantle obtained in Mexico. The third sample (A-GMEs1) consisted of ash obtained from combustion in the laboratory of sample GMEs1, a non-thoriated gas mantle obtained in Spain.

2.2. Measurement of ^{228}Th and ^{228}Ra activity concentrations

The activity concentrations of ^{228}Th and ^{228}Ra in the gas mantles were determined by gamma-ray spectrometry. The samples were measured in a spectrometer system consisting of a Canberra XtRa type high purity germanium (HPGe) detector, with a relative efficiency of 37% and a resolution of 1.77 keV for the 1.33 MeV photopeak of ^{60}Co. The detector was surrounded by 10 cm of lead shielding, lined with a 5 mm layer of copper. Additionally, the system featured an anti-coincidence device (an organic scintillator) to reduce background radiation levels. To determine the activity concentration of ^{228}Ra, the main gamma photopeaks of ^{228}Ac (338 and 911keV) were used. The activity concentration of ^{228}Th was determined using the weighted average emissions of ^{212}Pb (238.6 keV), ^{212}Bi (727.3 keV) and ^{208}Tl (583 and 2614keV), taking into consideration the corresponding branching ratios. Measurement times were longer than 3000 s to achieve a 95% confidence level, and the minimum detectable activities (MDAs) were in the range 0.1–0.4 Bq/g.

The detector efficiency calibration was performed by measuring the reference material IAEA-RGTh1 enclosed in the same type of container as that used for the samples. This reference material is a mixture of thorium ore and silicon powder with a known activity concentration. By using a reference material with the same radionuclides of interest, corrections due to coincidence effects are not necessary. Because of differences in composition, the bulk densities of the samples (0.10–0.33 g/cm^3) were much lower than those of the reference material (1.33 g/cm^3). However, no self-attenuation corrections were required because the effect of the matrix was negligible owing to the small sample size, the minimal thickness of the cylindrical geometry and the range of gamma energies of interest (238–2614 keV) [1].

The ash samples were measured in an anti-Compton gamma spectrometric system formed by a Canberra well-type HPGe detector surrounded by an NaI (Tl) well-type 5-inch detector, connected to six photomultiplier tubes for the anti-coincidence count. The whole system was surrounded by 10 cm of lead shielding to reduce background noise. The efficiency calibration for this anti-Compton system was performed by measuring the reference material IAEA-RGTh1, packaged in the same counting geometry as that of the ash. Measurement times were longer than 90 000 s to achieve a 95% confidence level. MDAs were in the range 0.15–0.35 Bq/g for the radionuclides of interest.

2.3. Measurement of ^{232}Th and ^{230}Th concentrations

The activity concentrations of ^{232}Th and ^{230}Th were determined by alpha-particle spectrometry. Aliquots ranging from 10 to 200 mg were taken, depending on the values obtained by gamma spectrometry. Samples were initially ashed and then alkali fused using $KHSO_4$ at 800°C for 20 min in accordance with the procedure described in Ref. [2]. After a period of cooling, the fused material was dissolved in 8M HNO_3, 10 mg of Fe^{3+} carrier was added and the precipitation of actinides was provoked by adding drops of concentrated ammonia until a pH of 9 was reached. In this way, the thorium isotopes became concentrated in the precipitate, which was separated from the supernatant by centrifuging. The thorium fraction was then diluted in 8M HNO_3 and subjected to a separation–isolation process based on liquid–liquid solvent extraction using tributylphosphate. Full details of the thorium isolation process are given in Ref. [3].

To prepare the alpha particle sources for measurement, the solutions with the thorium isolated fraction were electrodeposited onto stainless steel discs with an area of 15.2 cm^2. Electrodeposition was carried out in cells of polytetrafluorethylene with a platinum anode. The pH of the solution was adjusted to a value of 2.2 using ammonia vapour. The electrodeposition parameters were 1.5 A for 2 h. The alpha particle measurements were carried out using a Canberra Alpha Analyst system, equipped with passivated implanted planar silicon (PIPS) detectors of 450 mm^2 active area. The measurement time was 250 000 s, resulting in an MDA of less than 0.01 Bq/g for a 95% confidence level.

3. RESULTS AND DISCUSSION

The activity concentrations of ^{232}Th, ^{228}Ra, ^{228}Th and ^{230}Th measured in the gas mantles are shown in Table 1. The gas mantles obtained in Mexico were found to have ^{232}Th, ^{228}Th and ^{228}Ra activity concentrations of 482–683, 224–345 and 240–277 Bq/g, respectively, while the activity concentrations of ^{230}Th (a decay product of ^{238}U) were in the range 62–97 Bq/g. As expected, the gas mantles obtained in Spain were found to have very low activity concentrations of these radionuclides, ranging from below the MDA up to 1.37, 1.0, 0.9 and 0.41 Bq/g for ^{232}Th, ^{228}Ra, ^{228}Th and ^{230}Th, respectively. The following comments can be made with regard to the gas mantles obtained in Mexico:

(1) The ^{232}Th activity concentrations are much higher than the 10 Bq/g exclusion value for ^{232}Th, and the gas mantles therefore fall within the scope of international radiation protection standards.

(2) Assuming an average gas mantle mass of 2.9 g, the total ^{232}Th activities in the products are between 1398 and 1981 Bq — easily within the 350–4560 Bq range determined for thoriated gas mantles elsewhere in the world [4].

(3) The disequilibrium observed between ^{232}Th and its progeny ^{228}Th and ^{228}Ra is to be expected. The thorium used in the manufacture of the mantles has been chemically separated from the thorium mineral in which it was originally contained and only ^{232}Th and ^{228}Th would therefore be present in a newly manufactured gas mantle. As time passes, ingrowth of ^{228}Ra occurs, while the ^{228}Th in the gas mantle at any one time has two origins — the fraction originating from the ^{228}Th initially present in the mantle is decaying, while at the same time ingrowth of that same radionuclide is occurring as a decay product of the ^{228}Ra being generated. For gas mantles measured a few years after their manufacture, therefore, one can expect to find disequilibrium between ^{232}Th and its progeny ^{228}Ra and ^{228}Th, as evidenced by ^{228}Th:^{232}Th and ^{228}Ra:^{232}Th activity ratios less than unity.

(4) The concentrations of ^{230}Th found in the gas mantles correspond to a ^{232}Th:^{238}U ratio of about 5–10, consistent with the fact that significant amounts of uranium are found in minerals used as sources of thorium.

TABLE 1. RADIOACTIVITY IN GAS MANTLES

Sample code	Activity concentration (Bq/g)			
	Th-232	Ra-228	Th-228	Th-230
Thoriated gas mantles obtained in Mexico:				
GMx1	500 ± 24	310 ± 6	249 ± 10	62 ± 3
GMx2	554 ± 15	224 ± 6	259 ± 14	90 ± 5
GMx3	683 ± 22	320 ± 5	260 ± 15	85 ± 3
GMx4	665 ± 26	345 ± 4	277 ± 15	97 ± 5
GMx5	641 ± 29	343 ± 8	275 ± 15	77 ± 4
GMx6	482 ± 20	302 ± 7	240 ± 18	71 ± 4
Non-thoriated gas mantles obtained in Spain:				
GMEs1	<MDA	<MDA	<MDA	<MDA
GMEs2	1.37 ± 0.01	1.0 ± 0.1	0.9 ± 0.1	0.41 ± 0.01
GMEs3	<MDA	<MDA	<MDA	<MDA

Notes: MDA – minimum detectable activity.
The uncertainties quoted represent one standard deviation.

The activity concentrations of ^{232}Th, ^{230}Th, ^{228}Ra and ^{228}Th determined in the three gas mantle ash samples are shown in Table 2. The activity concentrations measured in sample A-MxFS (ash collected directly from street food stalls in Mexico) were very much lower than those measured in sample A-GMx6 (ash from the laboratory combustion of sample GMx6, a thoriated gas mantle obtained in Mexico). One possible explanation for this result is that the ash collected from the street food stalls might have been derived from a mixture of different brands of gas mantles that included both thoriated and non-thoriated products. As expected, the activity concentrations measured in sample A-GMEs1 (ash from the laboratory combustion of sample GMEs1, a non-thoriated gas mantle obtained in Spain) were by far the lowest, being all below the MDA. The following comments can be made regarding the ash originating from thoriated gas mantles:

(1) The ^{232}Th activity concentrations in the ash collected from the street food stalls, while very much lower than those in ash from the laboratory combustion of a gas mantle, imply some level of radiological risk to the public because this ash could be inhaled by nearby individuals or dispersed onto food heating plates.

(2) The ^{228}Ra:^{232}Th and ^{228}Th:^{232}Th activity concentration ratios determined in the laboratory-generated ash are very different from those determined in the gas mantle itself. This suggests that the behaviour of radium during the ashing process is different from that of thorium. This in turn affects the concentration of ^{228}Th, since it is a decay product of ^{228}Ra.

(3) All the results presented here, and particularly those relating to the use gas lanterns in street food stalls, highlight the need for regulatory control by the relevant Mexican authority of the use and disposal of gas mantles.

TABLE 2. RADIOACTIVITY IN GAS MANTLE ASH

	Activity concentration (Bq/g)			
	Th-232	Ra-228	Th-228	Th-230
Sample A-MxFS: Ash collected from street food stalls in Mexico	1.12 ± 0.04	0.38 ± 0.05	0.97 ± 0.1	0.22 ± 0.01
Sample A-GMx6: Ash from the laboratory combustion of thoriated gas mantle sample GMx6	1071 ± 72	2014 ± 47	1585 ± 37	118 ± 11
Sample A-GMEs1: Ash from the laboratory combustion of non-thoriated gas mantle sampleGMEs1	<MDA	<MDA	<MDA	<MDA

Notes: MDA – minimum detectable activity.
The uncertainties quoted represent one standard deviation.

ACKNOWLEDGEMENTS

The participation of the Radioisotope Service (CITIUS) at the University of Seville in this study is deeply acknowledged. C.D. Mandujano-García acknowledges the financial support of CONACyT (Grant: 328910) and the University of Guanajuato (Grant: CBM-05-2015-BMS-16) for a research stay at the University of Seville.

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Poster Presentation

LEAD-210 IN NORM FROM THE OIL AND GAS INDUSTRY

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Abstract

Oil, gas and associated water extracted from the Earth's crust has been in contact with rocks containing uranium and thorium for a long time and consequently contains appreciable amounts of radionuclides of natural origin in the ^{238}U and ^{232}Th decay series. These radionuclides are concentrated during the oil and gas production processes and cause scale formation within pipes. Topside equipment generally becomes contaminated with ^{210}Pb deposits, some of which are almost monomolecular and not detectable by visual inspection or, (because of the very low energy of the emitted gamma radiation) by using gamma spectroscopy.

1. INTRODUCTION

Geologists already knew about the presence of uranium, thorium and associated decay progeny in rock formations surrounding oil and gas reservoirs underground and have used them as a means for discovering their locations since the 1930s [1]. Most oil and gas resources occur in reservoirs formed by sedimentary rocks, particularly sandstones and limestones. As uranium and thorium are widely dispersed in crustal rocks, it is not surprising to find decay progeny of ^{238}U and ^{232}Th in the oil, gas and produced water that is brought to the surface. Significant amounts of NORM containing these radionuclides are generated during the exploration, production, maintenance and transport phases of the operation [2]. At many extraction and processing sites, ^{210}Pb (a beta and gamma emitter) and ^{210}Po (an alpha emitter) are found in mud tanks, pumps and drill pipes [3]. Both unsupported ^{210}Pb and supported ^{210}Pb can be transported through the production systems. The aim of this paper is to highlight, through a literature review, the two ^{210}Pb deposition mechanisms.

2. DEPOSITS CONTAINING ^{226}Ra AND ^{228}Ra

Changes in physico-chemical parameters caused by the introduction of water and in the temperature and pressure at extraction sites cause some of the ^{238}U and ^{232}Th decay progeny in the produced water to precipitate, forming radioactive deposits on the internal surfaces of pipes, pumps, valves, tanks and other equipment [4]. Locations of potential accumulation of these deposits in an offshore oil and gas production facility are shown in Fig. 1. [4]. When radionuclides and other minerals dissolved in the production water reach the surface, they co-precipitate, forming various waste deposits such as sediments in tanks and scale inside pipes. The scale contains compounds of barium, calcium and strontium, as well as radionuclides (mainly ^{226}Ra and ^{228}Ra), and precipitates as a result of changes in temperature and pressure of the produced water. As the oil in the reservoir becomes depleted, the amount of scale generated tends to increase. Reinjection of water into the reservoir to boost oil recovery further enhances the generation of scale.

3. DEPOSITS CONTAINING ^{210}Pb

Deposits generated in gas production facilities are rather different. Radon-222 (the immediate decay product of ^{226}Ra, with a half-life 3.8 d) decays quickly to ^{210}Pb with a half-life of 22 years (see Table 1). Since ^{222}Rn is a gas and is therefore highly mobile, its decay progeny are deposited at locations different from those associated with oil production scale. Internal surfaces of pipes and other equipment in gas processing plants are generally contaminated with ^{210}Pb rich deposits in the form of thin layers, some of which are not detectable by visual inspection.

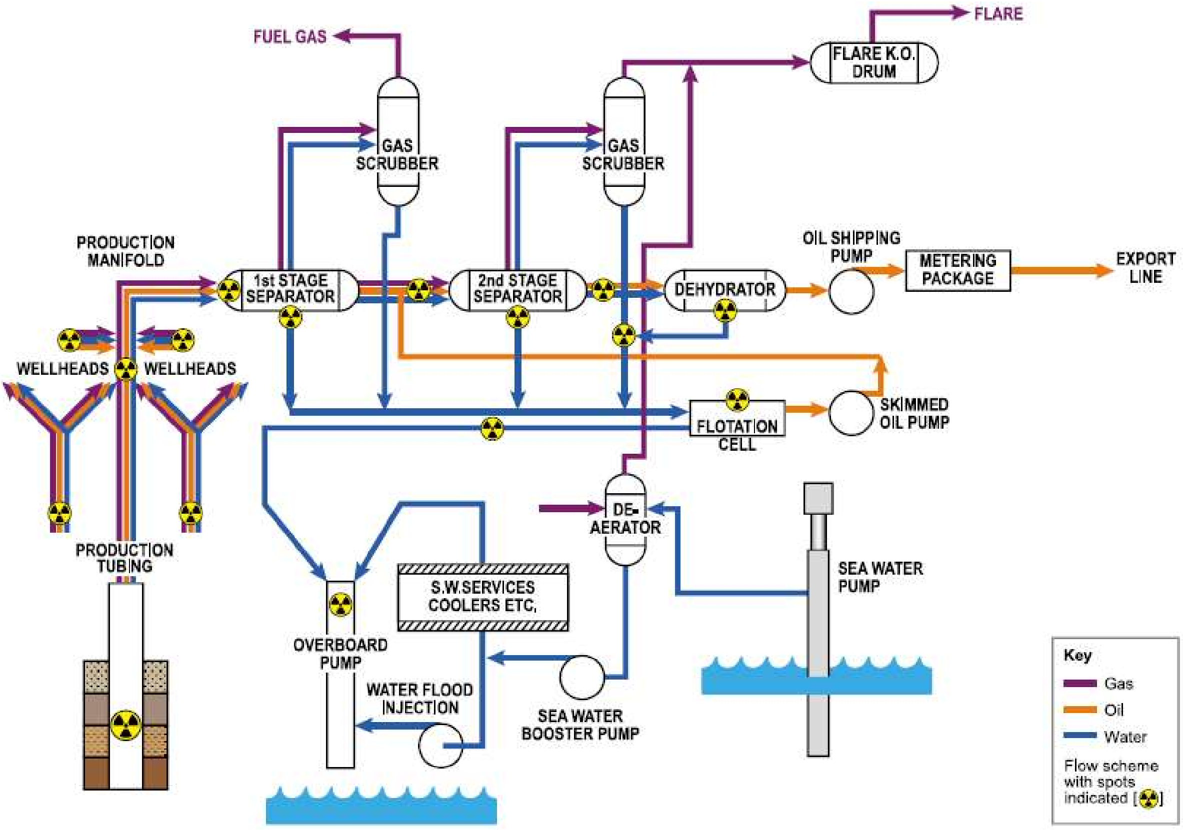
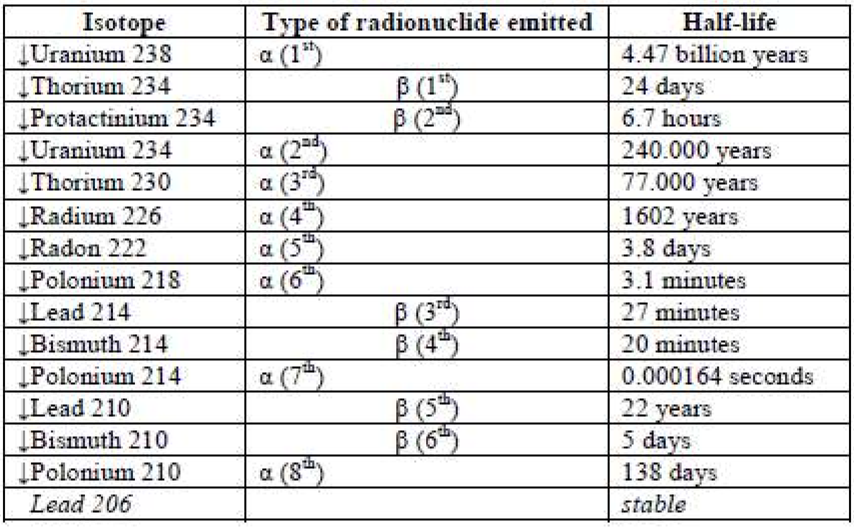


FIG. 1. Schematic view of an offshore oil and gas production facility showing potential locations of NORM accumulation.

TABLE 1. URANIUM-238 DECAY CHAIN.



Isotope	Type of radionuclide emitted	Half-life
↓Uranium 238	α (1st)	4.47 billion years
↓Thorium 234	β (1st)	24 days
↓Protactinium 234	β (2nd)	6.7 hours
↓Uranium 234	α (2nd)	240.000 years
↓Thorium 230	α (3rd)	77.000 years
↓Radium 226	α (4th)	1602 years
↓Radon 222	α (5th)	3.8 days
↓Polonium 218	α (6th)	3.1 minutes
↓Lead 214	β (3rd)	27 minutes
↓Bismuth 214	β (4th)	20 minutes
↓Polonium 214	α (7th)	0.000164 seconds
↓Lead 210	β (5th)	22 years
↓Bismuth 210	β (6th)	5 days
↓Polonium 210	α (8th)	138 days
Lead 206		*stable*

Both unsupported ^{210}Pb and supported ^{210}Pb can be transported through the system (see Fig. 2):

(1) Deposits containing unsupported ^{210}Pb are associated with dispersed particles and colloids, a portion of which are deposited on the inside surfaces of pipes forming a very thin and relatively stable deposit. Unsupported ^{210}Pb deposits have been observed in equipment that has been in contact with the produced water in such a way that, over time, the lead may be converted into other compounds, depending on the humidity of the gas in the presence of H_2S and other compounds [5, 6].

(2) Deposits containing supported ^{210}Pb are more likely to be found in places where the gas is retained for a time and thus in equipment that forms part of the gas treatment line, in which ^{222}Rn (and thus ^{210}Pb) traverses long distances through the pipeline before decaying and forming a deposit. The deposit takes the form of an ‘invisible’ film that contaminates the surface of the steel material [5]. The activity concentration of ^{210}Pb in the deposit can be above the exemption level specified international radiation protection standards. Besides being invisible, the deposit emits low energy gamma radiation, making in situ detection and evaluation difficult.

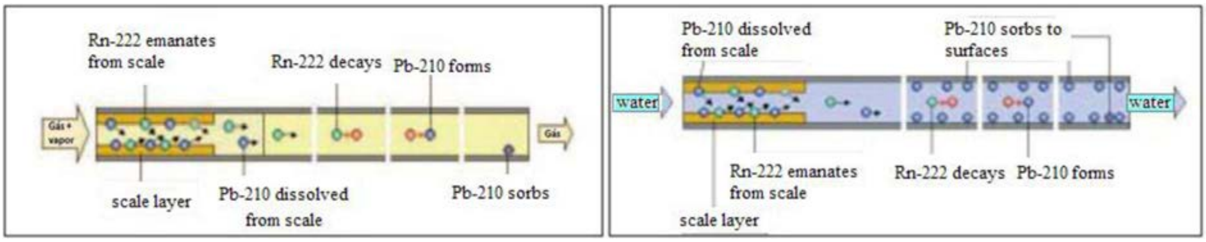


FIG. 2. Processes of supported ^{210}Pb and unsupported ^{210}Pb transport in pipelines.

4. CONCLUSIONS

Many references to the analysis of ^{210}Pb in NORM in the oil and gas industry can be found in the literature. However, with regard to the determination of ^{210}Pb supported by ^{222}Rn in the new context now approached, the references are practically non-existent.

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Poster Presentation

POTENTIAL FOR NORM WASTE IN SOME MINING AND PROCESSING INDUSTRIES OF UKRAINE

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Abstract

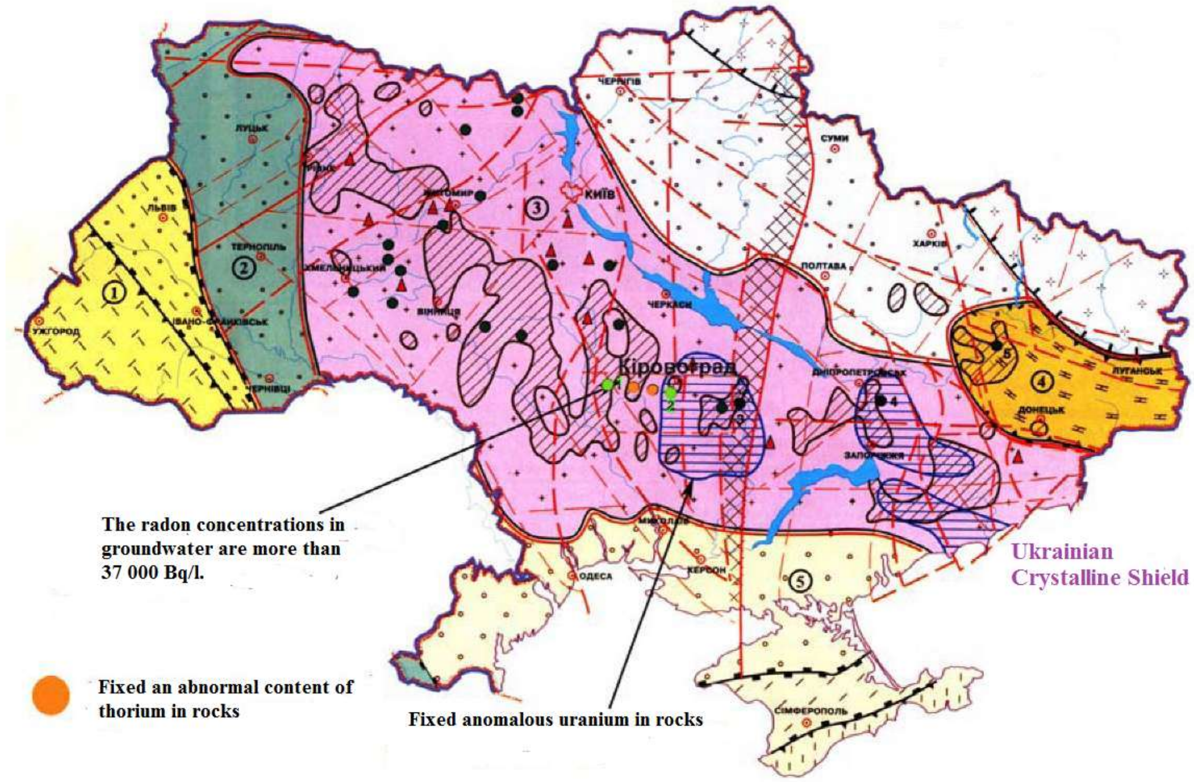
The mining and processing of minerals generates large amounts of waste containing radionuclides of natural origin. This study was conducted using gamma spectrometry. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb and ^{238}U were determined. The report includes the results for the following industries: oil and gas extraction, oil and gas refining, mining of non-ferrous metals, non-ferrous metal extraction industry, iron ore extraction industry. The conclusion drawn from the assessment is that only the oil and gas industry waste needs to be subject to regulatory control and thus falls within the definition of NORM. The activity concentrations in pipe sediments and silt from the oil and gas industry are as follows: ^{210}Pb: 3000–11 000 Bq/kg, ^{226}Ra: 2900–8400 Bq/kg, ^{232}Th: 800–2800 Bq/g, ^{238}U: 180–6900 Bq/g. It was established that the maximum activity concentrations in waste from the non–ferrous metal and iron ore extraction industries are ten times below the exemption levels for regulatory control and therefore do not fall within the definition of NORM.

1. INTRODUCTION

The mining and processing of metal ore minerals and oil may generate large volumes of waste containing radionuclides of natural origin. This waste includes mine tailings, smelter slag and oil extraction sludge, some of which contains elevated concentrations of uranium, thorium, radium and their decay products that were originally part of the process feed ore. In February 2015, the Cabinet of Ministers of Ukraine approved the implementation plan of the European Council Directive 2013/59/Euratom [1, 2]. Regulation of industrial NORM waste is one of the requirements in the Directive to be introduced in Ukraine. It should be noted that more than 45% of Ukrainian territory is situated on the ‘crystalline shield’ with high levels of natural radioactivity. Almost all Ukrainian mining and processing industries are located in the crystalline shield, namely: oil and gas extraction, oil and gas processing, mining of non-ferrous metals, the non-ferrous metal extraction industry and the iron ore extraction industry (Fig. 1). The purpose of this research work was to assess the activity concentrations of radionuclides of natural origin in industrial waste in Ukraine and assess which industries should be subject to regulatory control.

2. METHODS

Industries with potential NORM waste were approached in this research. Thirty two enterprises were the subject of investigation and 250 samples were taken in total. Sampling and analysis followed the same procedure in all cases.



The radon concentrations in groundwater are more than 37 000 Bq/l.

Ukrainian Crystalline Shield

Fixed an abnormal content of thorium in rocks

Fixed anomalous uranium in rocks

FIG.1. Geological structure of Ukraine.

2.1. Sampling

Sampling was done by industry representatives under supervision of inspectors of the State Nuclear Regulatory Inspectorate. The inspectors filled in the sampling protocols. The protocol contains information on the enterprise, its contact information, type of sample, date, and sampling coordinates. The waste samples were randomly taken at tailings sites, tailings ponds or, for oil industry enterprises, sludge separators.

2.2. Measurements

An ORTEK high resolution gamma spectrometry system with NaI (T1) scintillation detector and well were used for sample measurements. The samples to be measured were put inside the Marinelli container filling the space of the well with the detector crystal. By placing the sample in such way, a substantially higher geometric effectiveness of registration is achieved compared with a cylinder form crystals. A large volume well (150×150 mm) was used to provide sufficient sensitivity. An Amersham standard source was used for calibration of the gamma spectrometer. For the ‘Marinelli 1 litre’ geometry, the minimum detectable activity (MDA) values are 2 Bq/kg for ^{232}Th, 3 Bq/kg for ^{226}Ra and 10 Bq/kg for ^{40}K.

3. RESULTS AND DISCUSSION

3.1. Iron ore extraction industry

Waste from two iron ore production facilities, including prospecting sites and six adjacent mines, were investigated. At the first facility, it was established that ^{210}Pb accounted for over 40% of the total activity concentration in all samples, with a maximum activity concentration of 96 Bq/kg. The activity concentration of ^{226}Ra, ^{40}K and ^{238}U did not differ much among samples (14 Bq/kg, 21 Bq/kg, and 19 Bq/kg, respectively). The maximum concentrations were 5 Bq/kg for ^{232}Th and 1 Bq/kg for ^{235}U. At the second facility (four mines) the content of radionuclides in the tailings was established as follows: 68% ^{40}K (maximum activity concentration 499 Bq/kg) and 17% ^{210}Pb (maximum activity concentration 108 Bq/kg). The content of other radionuclides in the waste samples was 0.2% ^{235}U and 6.4 % ^{226}Ra, with maximum activity concentrations of 45 Bq/kg ^{226}Ra (mine no. 3), 26 Bq/kg ^{232}Th (mine no. 6), 43 Bq/kg ^{238}U (mine no. 4) and 1.5 Bq/kg ^{235}U (mine no.6). It was also established that the radionuclide concentrations in the waste depend on the year of production. Nevertheless, none of the samples exceeded the established exemption levels of the Council Directive 2013/59/Euratom.

A comparative analysis of the radionuclide content in mine tailings was performed as part of this work (Fig. 2). The ^{40}K concentration varied most in all the samples — by as much as 6 to 13 times in the samples from different mines. Other radionuclide concentrations varied by factors of 2–3 times between samples, except for ^{232}Th, for which the concentration could vary by as much as ten times. This could probably be explained by the geological characteristics of the mining blocks. Fig. 3 shows the average activity concentration of all NORM radionuclides in the waste for both of the facilities investigated.

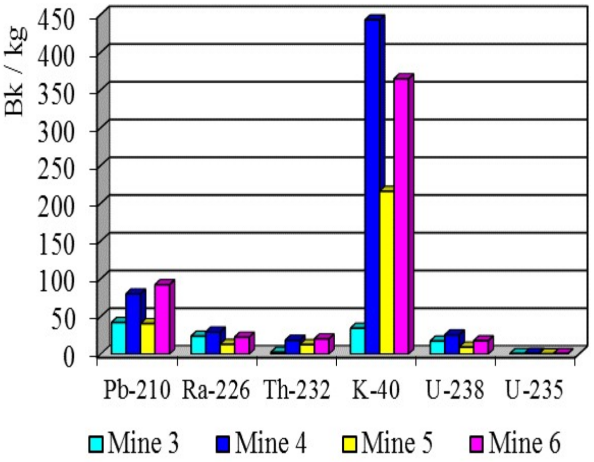


FIG. 2. Average radionuclide activity concentrations in waste from mines 3 to 6 in the second iron ore production facility.

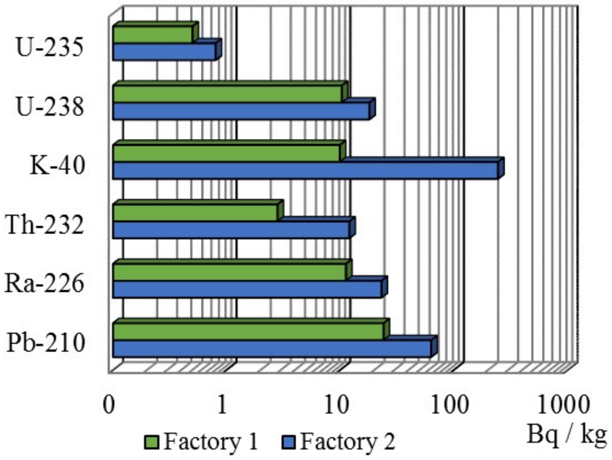


FIG. 3. Average radionuclide activity concentrations in waste from the two iron ore production facilities.

Based on the sample measurements and analysis it was established that the waste of these iron ore production activities does not need to be subject to regulatory control according to the European Council Directive 2013/59/Euratom and the IAEA International Basic Safety Standards (IAEA Safety Standards Series GSR Part 3) [3]. According to the established exemption levels, the radionuclide activity should be less then 1000 Bq/kg for ^{238}U and ^{232}Th decay chain radionuclides, and less than 10 000 Bq/kg for ^{40}K. For the first facility, the maximum activity concentration of uranium series radionuclides was ten times lower than the relevant exemption level, while the maximum ^{40}K concentration was 500 times lower than the relevant exemption level. For the second facility, the maximum activity concentration of uranium series radionuclides was ten times lower than the relevant exemption level, while the maximum ^{40}K concentration was 20 times lower than the relevant exemption level.

3.2. Steel industry

The steel industry in Ukraine uses local iron ore as well as metal scrap in the production processes. The industrial facilities may include mines, quarries and iron ore processing facilities. The radionuclide concentration in the waste depends on the applied technology and processes, as the radionuclide concentration in the raw materials does not vary much throughout the area. The radionuclide content of Ukrainian iron ore minerals is rather low, but in the case of processing, it would concentrate in the waste. The average concentrations of radionuclides throughout the industry are presented in Fig. 4. In total, four facilities were investigated in this research work. As expected, all the measured activity concentrations were below the exemption levels of the Council Directive 2013/59/Euratom.

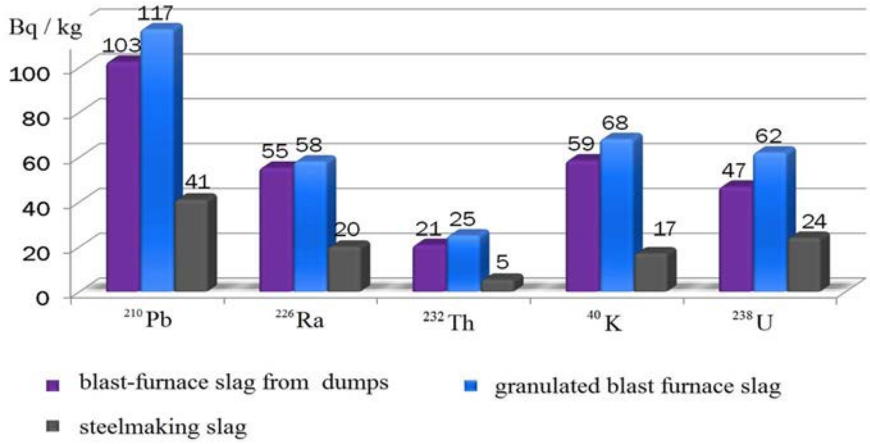


FIG. 4. Average radionuclide activity concentrations in waste from the steel industry.

3.3. Non-ferrous metal industry

Seven non-ferrous industrial facilities were investigated in this research programme. These included an alumina refinery, a titanium production facility, a ferroalloys facility and mineral fertilizer production facilities. The average radionuclide activity concentrations in the waste generated in these facilities are presented in Fig. 5.

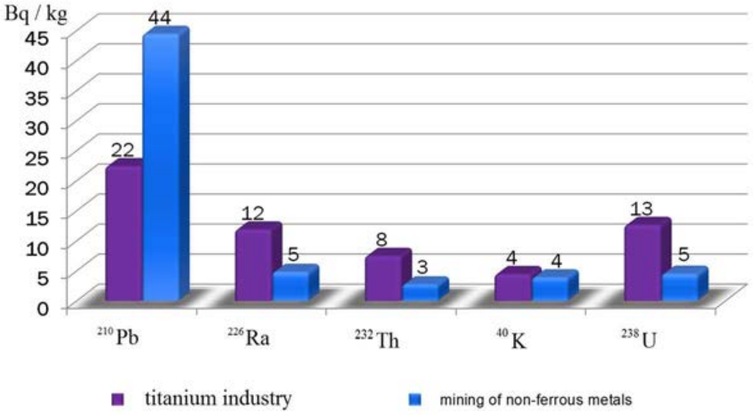


FIG. 5. Average radionuclide activity concentrations in waste from the non-ferrous metal industry.

From the results obtained, it can be concluded that the non-ferrous industry does not need to be subject to regulatory control requirements as all the samples were found to be significantly below the exemption levels.

3.4. Oil extraction industry

Six oil production and refinery facilities were investigated as part of this research. One facility was chosen for detailed investigation including gamma mapping of the industrial site. Oil spill spots and workplaces with elevated hazards were identified. High levels of gamma radiation were identified at oil spills, around barrels and at pipe storage sites. Several samples were taken of soil contaminated by oil spills and of pipe sediments and silt [4]. At the remaining five facilities, investigations were limited to the gathering of samples of pipe sediments and other waste. The results of this work are presented in Fig. 6.

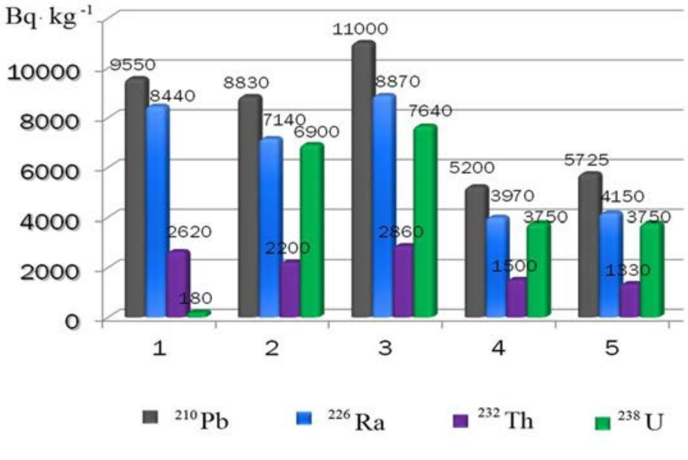


FIG. 6. Average radionuclide activity concentrations in waste from oil extraction facilities.

From the average activity concentrations measured in sludge and waste samples gathered at different locations within the industrial site, it was estimated that values might differ by as much as three times. The average activity concentrations for a given facility were up to 8870 Bq/kg for ^{226}Ra, 2860 Bq/kg for ^{232}Th and 11 000 Bq/kg for ^{210}Pb.

3.5. Oil refining industry

The oil refining industry in Ukraine consists of several major facilities and about 100 minor ones. In the course of the research programme, several major industrial sites were investigated and samples taken from the waste disposal ponds. It is worth mentioning that the sites have been in operation for over 50 years and significant amounts of waste have accumulated as a result. Fig. 7 shows the average radionuclide activity concentrations in the waste.

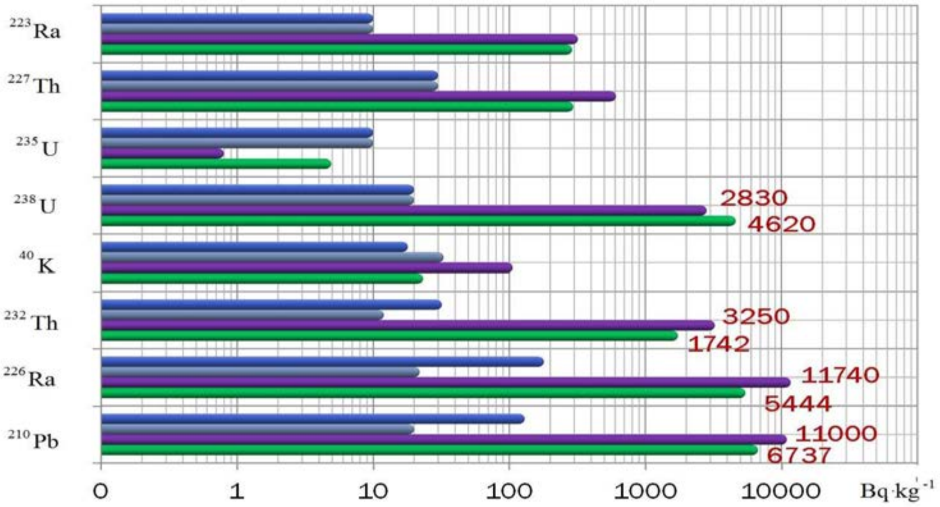


FIG. 7. Average radionuclide activity concentrations in waste from oil refining.

From the results of the sample measurements, the following average activities throughout the industry were obtained: ^{210}Pb: 6737 Bq/kg, ^{226}Ra: 5444 Bq/kg, ^{232}Th: 1742 Bq/kg and ^{238}U: 4620 Bq/kg. It is thus concluded that, because of the radioactivity levels in the waste, all oil extraction and processing sites in Ukraine should be subject to regulatory control.

4. CONCLUSIONS

(1) Monitoring of radioactivity concentrations in waste materials has identified the need to establish regulatory control of the oil industry in the country. The levels found in pipe sediments and silts at oil extraction facilities exceed the established exemption levels and range as follows: ^{210}Pb: 3000–11 000 Bq/kg, ^{226}Ra: 2900–8400 Bq/kg, ^{232}Th: 800–2800 Bq/kg, ^{238}U: 180–6900 Bq/kg.

(2) For regulatory control purposes, a package of regulatory documents must be developed.

(3) Further investigation and analysis of radionuclide concentrations in the oil extraction and processing industry is needed.

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OCCUPATIONAL RADIATION PROTECTION AND NORM METROLOGY

(Session 7)

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UMEX: A WORLDWIDE IAEA SURVEY OF OCCUPATIONAL EXPOSURE IN FACILITIES FOR THE PRODUCTION OF URANIUM

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Abstract

The production of uranium is carried out in nearly 20 countries and employs tens of thousands of workers. The IAEA Uranium Mining Exposure (UMEX) project involved an international survey of occupational exposure in facilities for the production of uranium in 2012. The survey accounted for more than 85% of worldwide uranium production. The results were analysed according to the types of work carried out, the production stages and the exposure pathways. Further analysis was performed through the use of dose distributions for the various facilities and selected groups of workers. Issues around inconsistencies in dose calculation and aspects such as background subtraction were explored. The overall findings showed that the industry was in compliance with international radiation protection standards and, in particular, was strongly committed to the optimization of protection.

1. INTRODUCTION

With the current level of interest in nuclear power, there has been an increase in uranium exploration and in the development of new uranium production facilities in many countries. Such facilities include in situ leaching operations and facilities for the mining and processing of uranium ore. World production in 2012 was 58 344 t of uranium[1], emanating from approximately 50 production facilities in nearly 20 countries. Workers engaged in uranium production receive external exposure to gamma radiation emitted from uranium ore, process materials, uranium products, tailings and other process residues. In addition, they receive internal exposure from the inhalation of airborne dust particles containing long-lived alpha activity and from the inhalation of radon and its short-lived decay progeny. The number of uranium production workers may increase substantially over the next few years.

Against this background, the IAEA has established the Uranium Mining Exposure (UMEX) project. The general aim of the project is to strengthen and enhance the radiation protection of uranium production workers, while more specific aims are to increase the opportunities for optimization of protection and to support quality assurance programmes across the industry. Within the framework of the project, the following key activities have been initiated with respect to uranium production workers worldwide:

(a) Development of an information system for occupational exposure;
(b) Evaluation of the current occupational radiation protection situation;
(c) Identification of instances of good practice, opportunities for improvement and, where appropriate, actions to be implemented for assisting employers, workers, regulatory

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[1] Data from World Nuclear Association, London, 2012.

bodies and other stakeholders in implementing the principle of optimization of protection and safety.

In 2012, the IAEA developed a questionnaire which was distributed to uranium producing countries. In 2013, responses to the questionnaire were received from 36 operating facilities which, between them, accounted for about 85% of worldwide uranium production. This paper presents:

(a) The results of the information survey and a preliminary analysis thereof;
(b) A summary of current practices for monitoring and reporting of occupational exposure;
(c) A summary of occupational exposures reported for 2012.

2. MONITORING AND DOSIMETRIC PRACTICES

2.1. External exposure to gamma radiation

2.1.1. Monitoring techniques and strategies

The use of various monitoring techniques and strategies is summarized in Figs 1 and 2. Most facilities used personal monitoring of all occupationally exposed workers using thermoluminescence dosimeters (TLDs). In a relatively small number of facilities, individual monitoring was restricted to selected individuals or to workgroup averaging. A few facilities used area monitoring or a combination of area monitoring and personal monitoring.

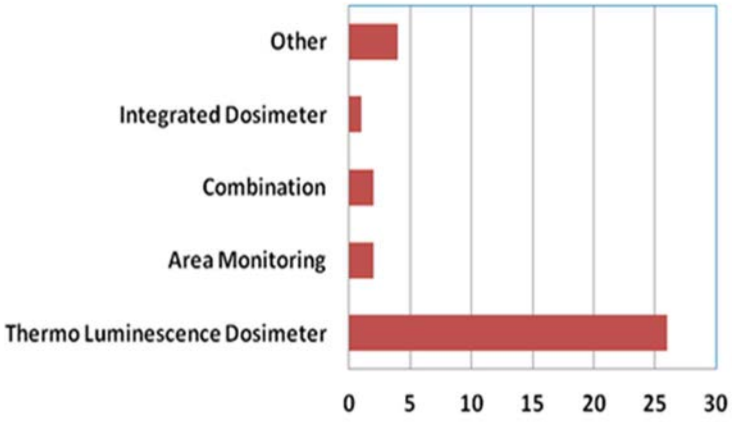


FIG. 1. Use of monitoring techniques for assessment of external exposure to gamma radiation (number of facilities).

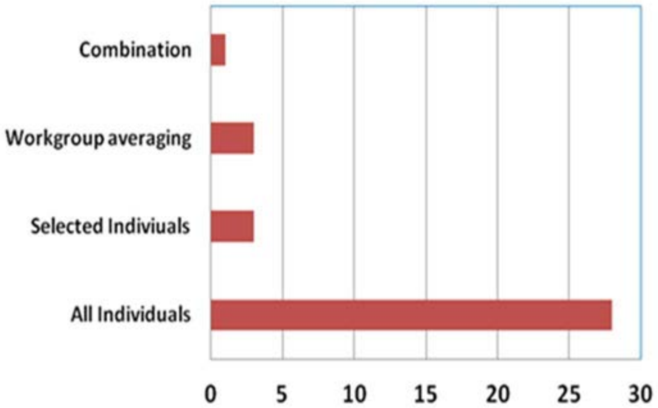


FIG. 2. Use of monitoring strategies for assessment of external exposure to gamma radiation (number of facilities).

Devices such as TLDs measure not only the occupational exposure but also the exposure due to background gamma radiation and exposure during transit. Therefore, the occupational contribution will be overestimated unless control badges or other correction methodologies are used. This overestimation will depend on the location but is likely to be of the order of 1 mSv/a. It is evident from Fig. 3 that less than 50% of the facilities subtracted the background gamma exposure from the monitoring result, implying that the exposures at the remaining facilities may have been significantly overestimated.

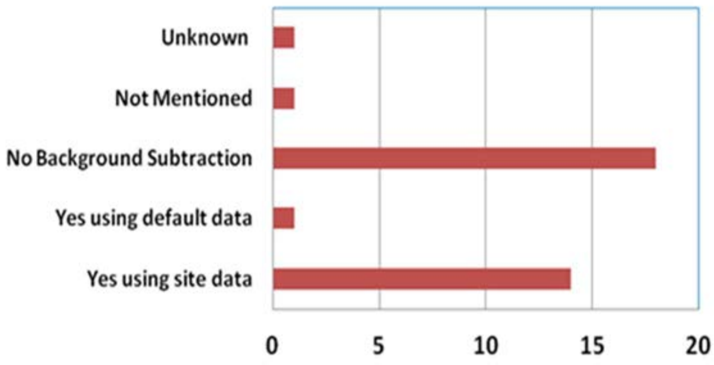


FIG. 3. Use of background subtraction for assessment of external exposure to gamma radiation (number of facilities).

2.1.2. Dosimetric aspects

Owing to the predominant use of TLDs, the dosimetric characteristics of the gamma radiation are closely aligned with the characteristics of the TLD. It is assumed that the data used would be Hp(10) equivalent but this was not defined in the survey.

2.1.3. Common and divergent aspects

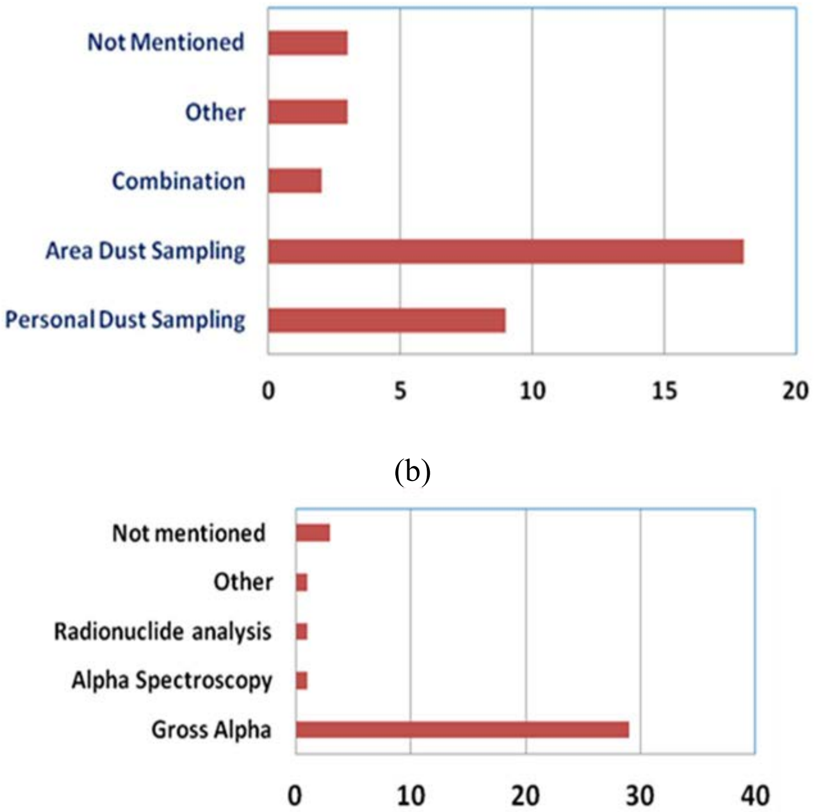
The predominant use of individual monitoring using TLDs provided a high level of consistency between facilities. On the other hand, the approximately equal split between facilities that used background subtraction and those that did not represented a major source of inconsistency, although this is unlikely to be significant in terms of demonstrating regulatory compliance.

2.2. Internal exposure due to inhalation of long-lived radionuclides in airborne dust

2.2.1. Monitoring methods

The monitoring methods used are summarized in Fig. 4. Personal dust sampling and area dust sampling were both widely used for the collection of airborne dust samples. In each case, the samples were usually analysed using gross alpha counting. Only two facilities used a more precise technique (alpha spectrometry or radionuclide analysis) in order to account for the isotopic composition of the dust (which could be important for dosimetry). Although not noted in the questionnaire responses, it was assumed that the dust samplers were size selective and that the dust measurements reflected respirable dust only. Since the background exposure to long-lived radionuclides in airborne dust is very small, its effect on the measurement of occupational exposure was regarded as insignificant.

(a)



(b)

FIG. 4. Use of methods for determining long-lived radionuclide activity concentration in air (number of facilities); (a) Dust collection, (b) Activity measurement.

Variations in exposure with time were handled using one of three monitoring strategies: workgroup averaging, periodic monitoring and permanent individual monitoring, as shown in Fig. 5. The strategy described as permanent individual monitoring included the continuous use of individual dosimeters for other sources of exposure (i.e. gamma radiation and short-lived radon decay progeny) into which a dust sampling capability had been incorporated.

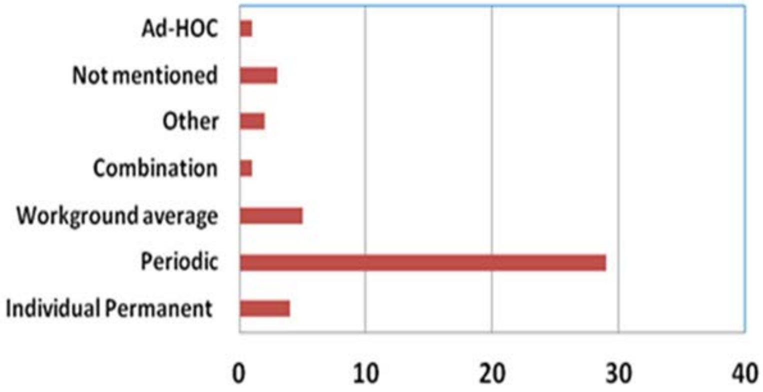


FIG. 5. Use of monitoring strategies for assessment of internal exposure to long-lived radionuclides in airborne dust (number of facilities).

2.2.2. Dosimetric aspects

As indicated in Fig. 6, internal dosimetric measurements such as direct bioassay were not widely used. However, six facilities used the assessment of uranium in urine for dosimetry purposes, together with an additional biological monitoring technique[2] in the event of an incident.

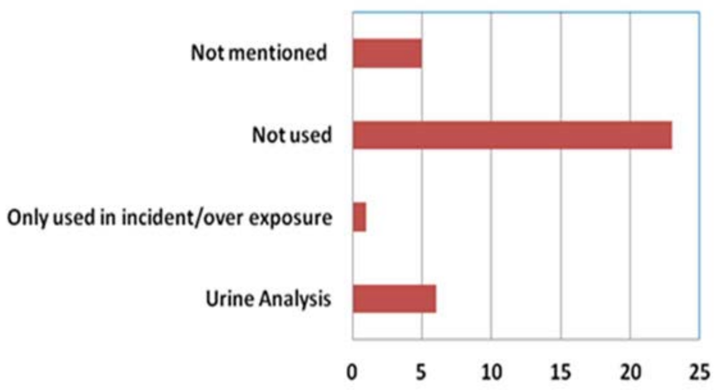


FIG. 6. Use of bioassay techniques for assessment of inhalation dose from long-lived radionuclides in airborne dust (number of facilities).

When personal dose sampling, workgroup averaging or area averaging methods are used, the occupancy time is needed for estimating the annual effective dose. As shown in Fig. 7, timesheets were the primary means of determining the occupancy time although there was significant use of electronic measurement techniques, reflecting the general trend in this direction. If timesheets are used for dosimetric purposes, then every effort should be made to ensure their accuracy in order to minimize errors in calculating the dose.

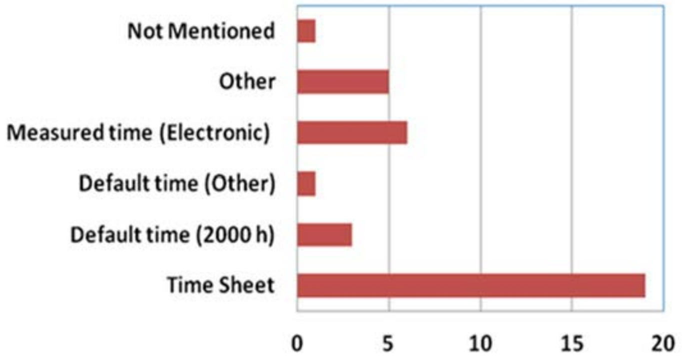


FIG. 7. Use of methods for determining occupancy time for assessment of inhalation dose from long-lived radionuclides in airborne dust (number of facilities).

While most facilities used dose conversion factors specified by the regulatory body or by international recommendations and standards, some used default values based on experience, measured values or the most conservative values. As expected, there was a wide range in the reported dose conversion factors for inhaled long-lived alpha activity. This can be attributed to differences in the composition of the material. For example, uranium ore is likely to be in equilibrium, tailings will be depleted in uranium and the final product is essentially pure uranium. Even for the final product, the reported dose conversion factors apparently varied by more than an order of magnitude. This may be due to specific studies at some facilities of airborne particle characteristics such as particle size and solubility. The dose conversion factor may also depend on assumptions related to certain parameters such as the fraction of radon progeny retained in a collected dust sample and hence the numbers of long-lived alpha particles contributing to the direct alpha measurement. A few facilities included the contribution of the uranium-235 decay series radionuclides to the total dose.

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[2] Biological monitoring potentially includes a range of techniques, including urine analysis, faecal sampling, radon exhalation in breath and chest or whole body counting.

2.2.3. *Common and divergent aspects*

The most significant variation in the calculation of the dose from long-lived radionuclides in airborne dust was in the dose conversion factors. The wide range of values used would have a significant influence on the calculated dose. In particular, three facilities included a very low dose conversion factor for non-calcined ammonium diuranate (ADU), although its use in practice was unconfirmed. In the facilities in question, the dose conversion factor had only a small impact on the reported dose owing to the limited potential for exposure to airborne dust.

2.3. Inhalation of short-lived radon progeny

All facilities measured radon or its short-lived decay progeny for occupational exposure assessment and dose estimation. The use of various monitoring techniques and approaches for assessment of exposure is shown in Figs. 8 and 9.

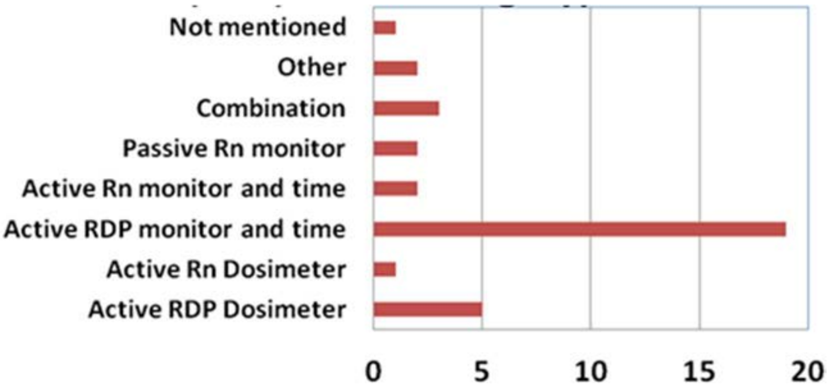


FIG. 8. Use of monitoring techniques for assessment of internal exposure to short-lived radon decay progeny (number of facilities); (Rn: Radon; RDP: short-lived radon decay progeny).

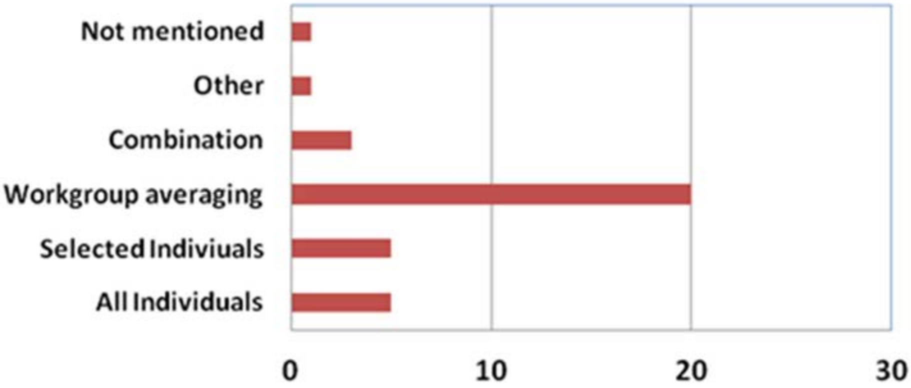


FIG. 9. Use of monitoring strategies for assessment of internal exposure to short-lived radon decay progeny (number of facilities).

3. NUMBERS OF OCCUPATIONALLY EXPOSED WORKERS

The numbers of occupationally exposed workers at each facility are shown in Fig. 10. The following should be noted:

(a) Operation 2 dominates the worker numbers with a figure of more than 10 000. However, the dose distribution for this particular facility indicated that approximately 75% of the workers received annual doses less than 0.5 mSv, suggesting that such workers might not have been engaged in work that involved close proximity to radioactive material.

(b) Operation 13 is an amalgamation of data for 15 facilities which report though a single organization. It is therefore included in subsequent data analysis as 15 different facilities so as to more correctly reflect the significance of this particular data set.

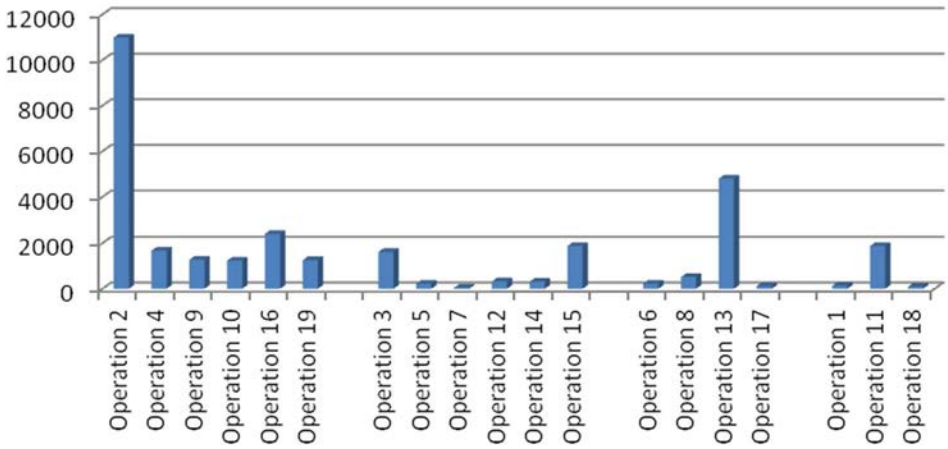


FIG. 10. Number of occupationally exposed workers, by facility.

4. ANNUAL EFFECTIVE DOSE

The annual effective doses estimated for each facility are shown in Fig. 11. The average across all facilities, weighted according to the number of occupationally exposed workers at each facility, was less than 5 mSv. One facility (Operation 3) reported a maximum annual dose of about 30 mSv. Further investigation revealed that the dose was due entirely to gamma exposure of a worker in the final product recovery section in one quarterly TLD issue period. It is suspected that the measurement was erroneous as a result of an incorrectly exposed TLD, erroneous reporting or some other non-operational problem. If this one result were to be discarded, the maximum annual dose for that facility would have been less than 10 mSv. For all other operations, the maximum annual effective dose was less than 10 mSv, except for one facility (Operation 4) at which the maximum dose was about 15 mSv — this dose included contributions from all three exposure pathways and was considered to be a valid result.

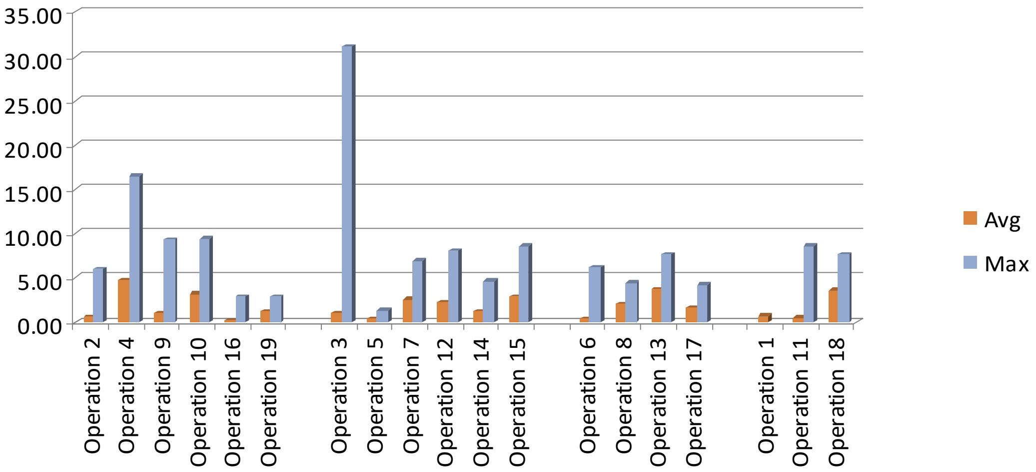
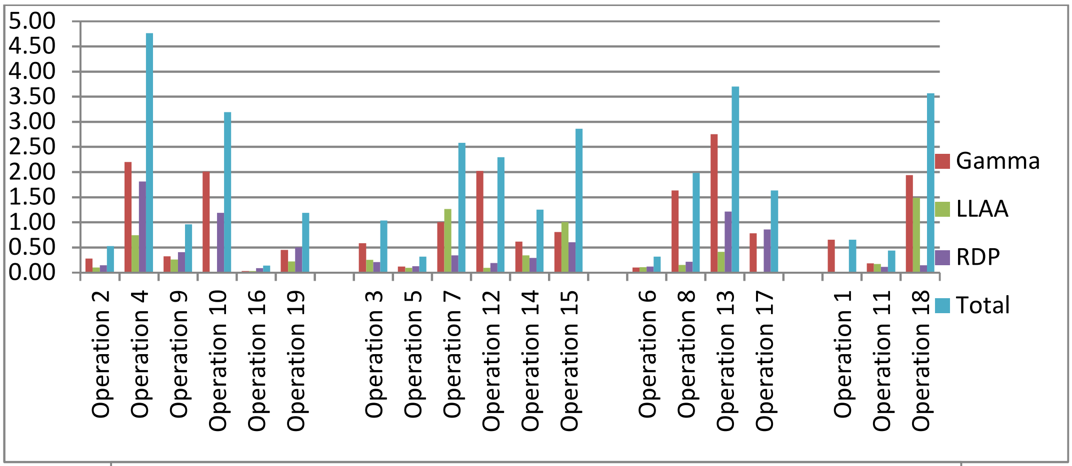
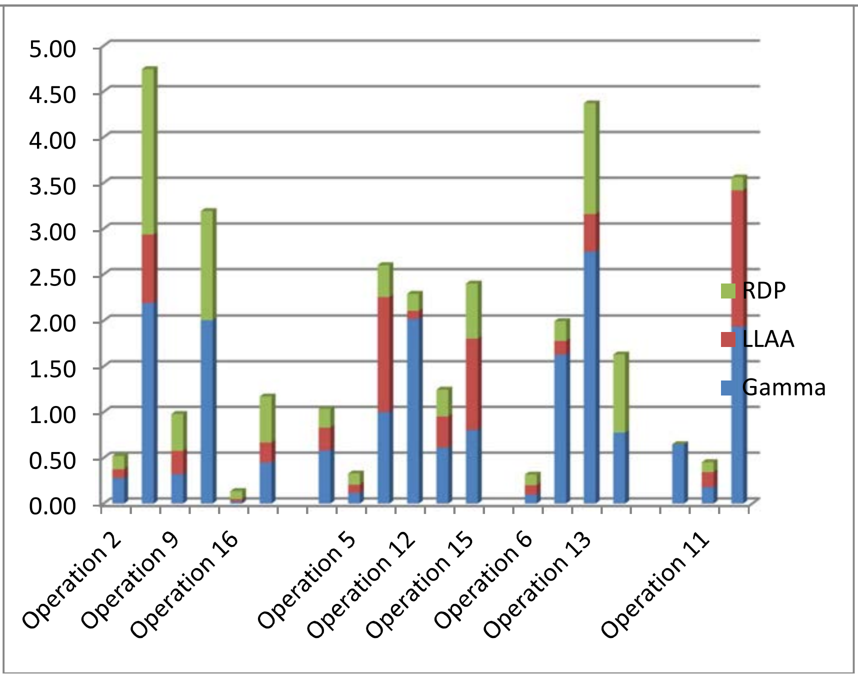


FIG. 11. Annual effective dose (in millisieverts), by facility.

The total annual dose from each facility, together with the contributions from the three exposure pathways are shown in Figs 12 and 13. There were considerable variations between facilities. While most facilities provided data for all three pathways, Operation 10 did not provide any information on the inhalation of long-lived radionuclides in airborne dust and Operation 1 provided only the gamma dose. Operation 13 (which represented 15 separate facilities) did not use background subtraction for gamma exposure. As explained in Section 2.1.1, the gamma dose contributions could therefore have been overestimated by about 1 mSv.





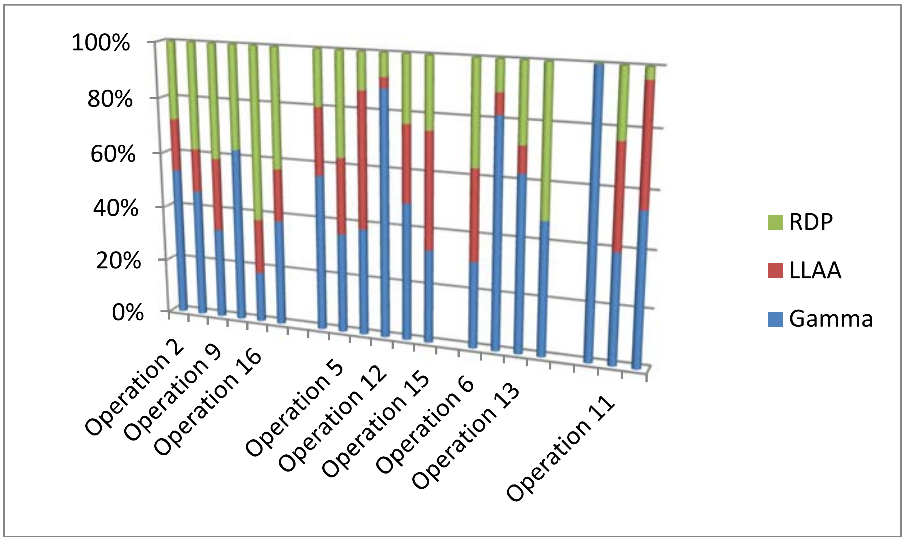
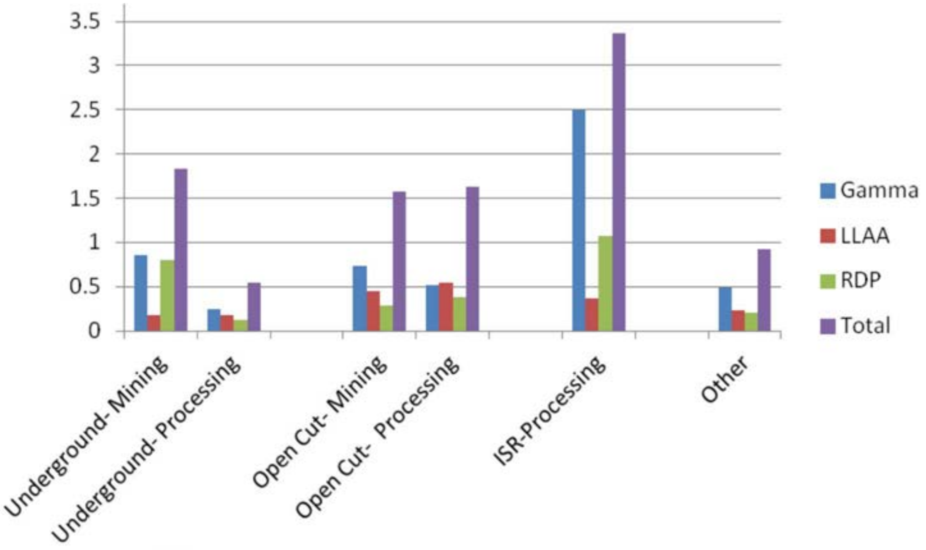
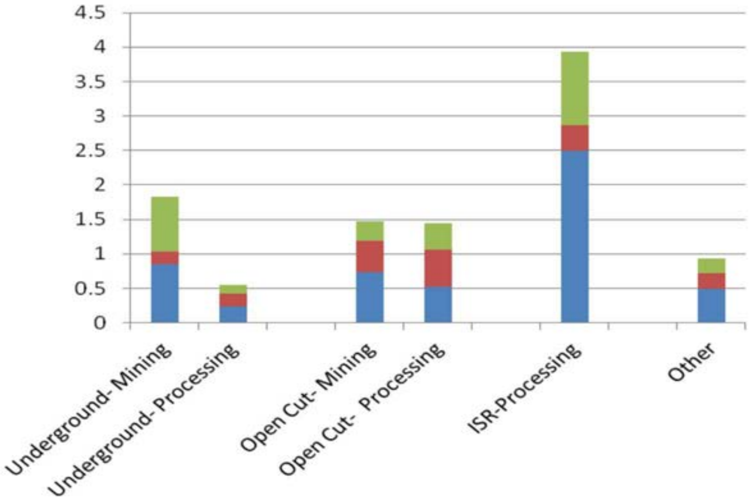
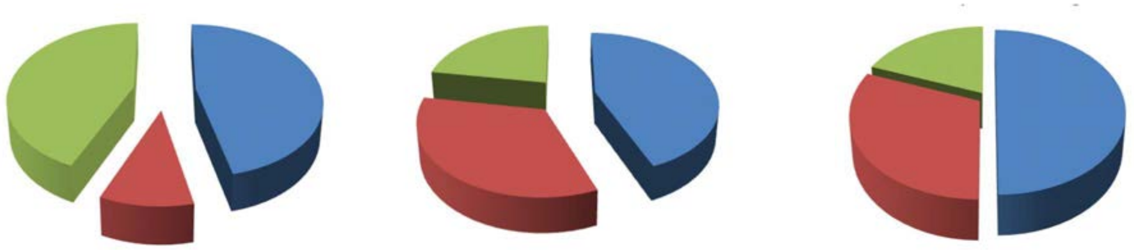


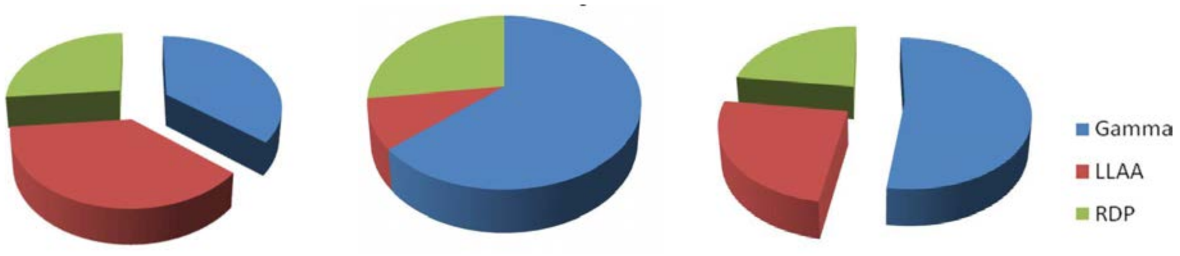
FIG. 12. Total annual effective dose and contributions from each exposure pathway (in millisieverts and percentage), by facility (LLAA: long-lived alpha activity; RDP: short-lived radon decay progeny).







Underground extraction: mining Underground extraction: processing Opencast extraction: mining



Opencast extraction: processing In situ recovery Other

FIG. 13. Total annual effective dose and contributions from each exposure pathway (in millisieverts), by type of uranium production activity (LLAA: long-lived alpha activity; RDP: short-lived radon decay progeny; ISR: in situ recovery). The category 'other' included uranium recovery from rehabilitation, waste water treatment and contract processing.

5. ANALYSIS OF RESULTS

5.1. Total annual effective dose

The following can be noted from the results presented in Figs 12 and 13:

(a) For mining and processing facilities at sites using underground extraction, the overall dose was heavily influenced by Operation 2 owing to the large number of occupationally exposed workers recorded for this particular facility (see Fig. 10).
(b) Among the mining and processing facilities at sites using opencast extraction, Operations 3 and 15 had by far the highest numbers of occupationally exposed workers and therefore had a major influence on the overall dose.
(c) For facilities using in situ recovery, Operation 13 had the highest number of occupationally exposed workers but, because this was an amalgamation of 15 facilities which were treated separately in the weighted averaging process, the overall dose was not unduly influenced and was therefore considered representative of in situ recovery facilities in general.

5.2. Contributions from the three exposure pathways

The following can be noted from the results presented in Figs 12 and 13:

(a) In underground mines, the contributions from external gamma exposure and internal exposure to inhaled short-lived radon decay progeny were similar (47% and 43%, respectively), while the contribution from the inhalation of long-lived radionuclides in airborne dust was much smaller (10%). This reflects the approach taken in modern underground mines, namely, a combination of dust suppression, good ventilation and shielding against gamma radiation.

(b) In the processing of ore derived from underground mining, the contributions from external gamma exposure and internal exposure to inhaled long-lived radionuclides in airborne dust were similar (44% and 34%, respectively), while the contribution from internal exposure to inhaled short-lived radon decay progeny was smaller but still significant (22%). However, since background subtraction was not generally used for exposure to radon decay progeny, a significant proportion of this contribution may not have been related to the ore processing operation.

(c) In opencast mining operations, the main contribution was from external gamma exposure, as would be expected for modern mining methods. Gamma shielding is not generally possible beyond that provided by the heavy earthmoving equipment that many workers operate. The next most significant contribution was that from the inhalation of long-lived radionuclides in airborne dust — this was dominated by Operations 3 and 15 which were both in semi-arid regions where airborne dust was likely to be more prevalent because of less water being available for dust suppression and more rapid drying of material. The inhalation of short-lived radon decay progeny was the least significant exposure pathway, as would be expected given the natural dispersal of radon within large open pits.

(d) In the processing of ore derived from opencast mining, the relative contributions to the total dose were, as expected, similar to those in facilities processing ore from underground mining (see (b) above).

(e) In operations involving in situ recovery, the main contributor to the total dose was external exposure to gamma radiation. It should be noted, however, that this result is almost totally related to Operation 13 which is an amalgamation of 15 separate facilities, none of which applied a correction for background gamma radiation, leading to the likelihood of a significant overestimation of occupational exposure from this pathway.

(f) Facilities categorized as ‘other’ included facilities for uranium recovery from rehabilitation, waste water treatment and a form of contract processing known as ‘toll milling’. The relative contributions from the three exposure pathways varied widely. Operation 1 provided only gamma exposure data and this pathway was therefore recorded as the only contributor to the total dose. In the case of Operation 11, a contract processing operation which had the largest number of occupationally exposed workers, external gamma exposure was the largest contributor to the total dose — this was expected given the nature of a purely contract processing operation.

5.3. Dose distributions

Most of the facilities provided data on the distribution of doses across groups of workers. By examining the number of workers in each group and the extent to which the data were clustered around the low doses, it was possible to determine the degree to which a group of workers was representative and whether there were potential outliers or individuals significantly different from the group. The dose distributions for the various facilities are shown in Fig. 14. Operation 2 was characterized by a large number of workers, of which more than 87% received an annual effective dose of less than 1mSv. This implied that the data were not focused on workers with a greater potential for exposure (for instance, designated radiation workers) but was a record of all workers who entered areas of the operation that were subject to some form of control. This approach does ensure that all potential exposures are recorded but also biases the average data to very low doses. For other operations, the data were focused on much smaller groups of workers and the doses were clustered at higher levels of exposure. This implied that the recorded data were more heavily focused on designated radiation workers and hence would have a higher average. Although both these approaches are valid, the importance of understanding the nature of the data is highlighted — simply using averages can give an incorrect interpretation of the relative radiological impacts.

By normalizing the distribution (i.e. dividing the number of workers in each dose range by the total number of workers) it is possible to examine and compare the data more directly. The normalized dose distribution data are shown in Fig. 15. All facilities appear to have a dose distribution with the characteristics of a log-normal distribution, in which higher numbers of workers are clustered around the lower doses. Because some operations appeared to have used different dose ranges (e.g. 1 mSv instead of 0.5 mSv) care must be taken in analysing the data. However there appears to be two distinct groupings, with some facilities having a dose distribution peaking in the very low dose range (0–0.5 mSv) and others having a dose distribution peaking in higher dose ranges. There are three possible reasons for this:

(i) The data provided were based on dose ranges wider than 0.5 mSv;
(ii) The selection of workers to be monitored concentrated on workers with the potential for higher exposures, shifting the distribution upwards;
(iii) The data recorded included natural background exposure (particularly for gamma monitoring) and therefore included a background contribution of up to 1mSv.

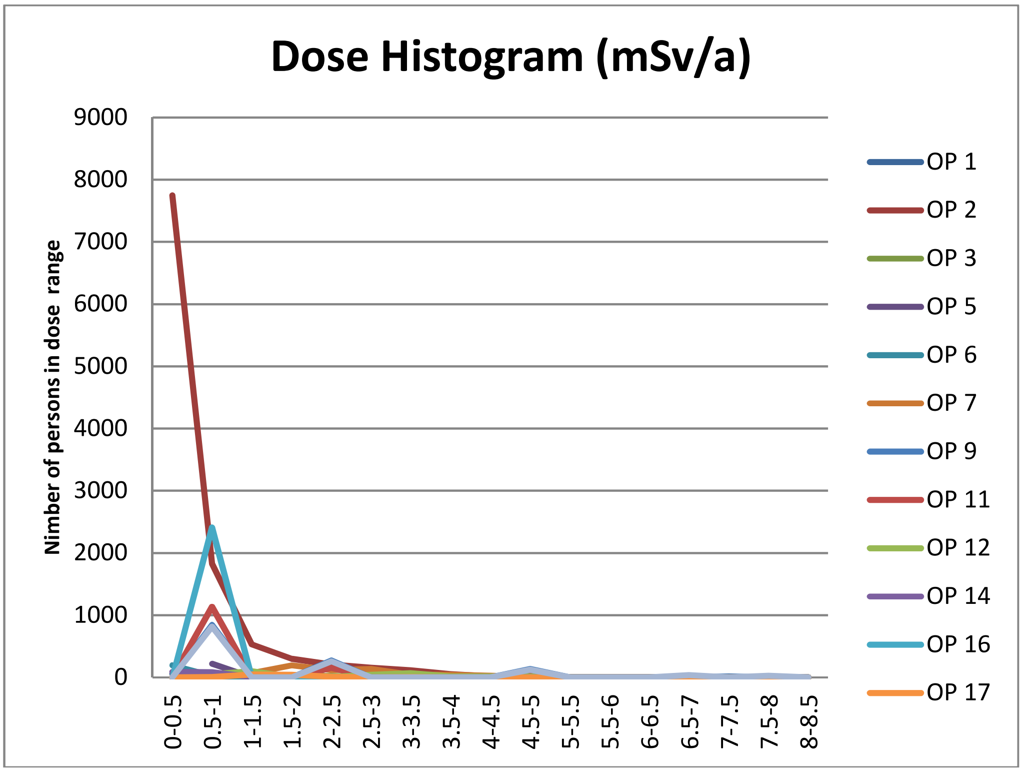


FIG. 14. Distribution of annual effective dose (in millisieverts) received by workers at various facilities.

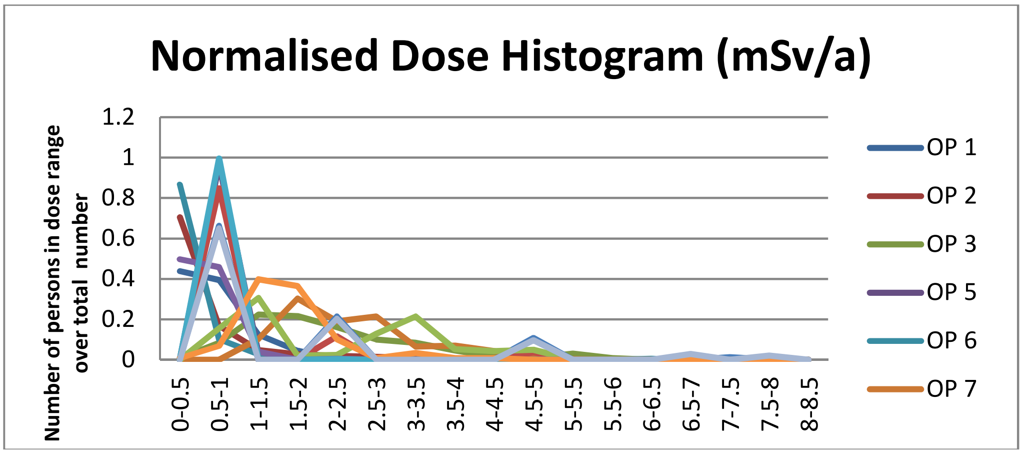


FIG. 15. Normalized distribution of annual effective dose (in millisieverts) received by workers at various facilities.

The dose data can also be utilized for examining more specific workgroups within a facility. For a workgroup which is representative of all its members, the resulting distribution generally follows a log-normal distribution. However, it is common for a workgroup to be a combination of two or more sub-groups, leading to a bimodal or multimodal distribution. For example, a selection of workgroups was analysed from Operation 2. The results, presented in Fig. 16, show that the distributions for the electricians and, to a lesser degree, the production drillers are characteristic of groups which are internally consistent. However, the distributions for the ore handlers and the raise drillers show evidence of a bimodal distribution. This is not unexpected if the range of work for the group is considered. For example, raise drilling operations can often be split into various tasks, some of which have a higher potential for exposure. If some individuals in the workgroup concentrate on only one task (for instance, controlling the drill rather than being at the face changing the rods) then this bimodal distribution could be expected. Similarly, workgroups often incorporate some individuals who, according to their job requirements, will have a different potential for exposure than others in the group. The most common example would be for the shift supervisors who may move between operations and be doing more administrative duties and hence could be expected to have a lower potential for exposure.

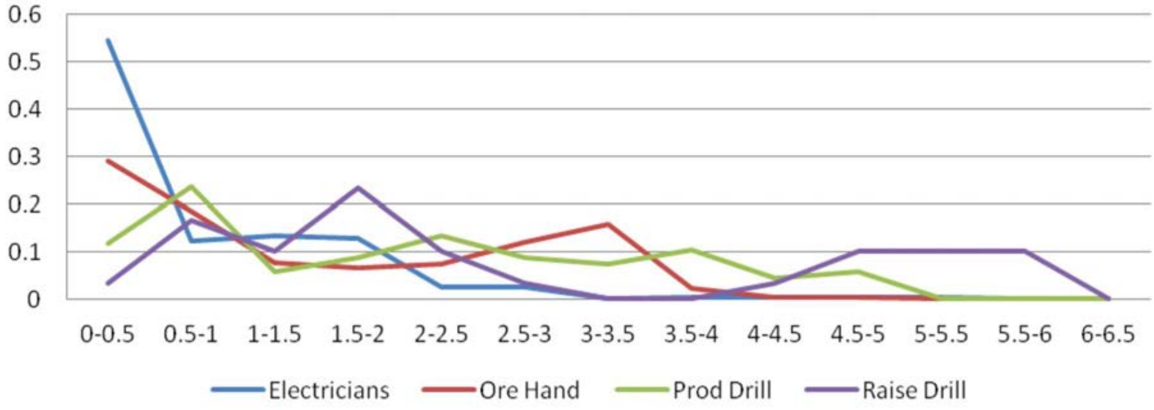


FIG. 16. Normalized dose distributions for selected workgroups.

The dose distribution is an underutilized tool for the interpretation of dose data. At the very minimum, knowledge of the nature of the dose distribution is necessary for the understanding and correct interpretation of parameters such as the mean and standard deviation.

It is also a useful tool for determining whether the members of a workgroup are similarly exposed or whether there are members with a significantly different exposure profile. As the use of workgroup averaging is a common tool in dose assessment, this understanding of the internal consistency of a workgroup is a means for improving the accuracy of the dose assessment.

6. MAIN OBSERVATIONS AND CONCLUSIONS

A worldwide survey of occupational radiation exposure in the production of uranium was performed in 2013. Responses were received from 36 operating facilities covering nearly 85% of global uranium production. A review of information from the responses to the UMEX questionnaire has identified several observations of a general nature as well as more specific observations on assessments related to individual exposure pathways. These are summarized as follows:

(a) General observations:
   (i) Although several methods have been adopted for the production of uranium, the most widely used method is in situ leaching, followed by underground and opencast mining of uranium ore;
   (ii) The most widely used technique for the processing of uranium ore is acid leaching, followed by alkaline leaching;
(b) Assessment of external exposure to gamma radiation:
   (i) Most facilities use TLD methods for the assessment of individual doses;
   (ii) The most widely used assessment strategy is the monitoring of all occupationally exposed workers, followed by the monitoring of average exposures of selected groups and the monitoring of selected individuals;
   (iii) Approximately 50% of facilities do not use background subtraction, leading to an overestimation of the doses received by workers;
(c) Assessment of internal exposure from the inhalation of long-lived radionuclides in airborne dust:
   (i) Approximately 50% of facilities use workplace dust sampling and 50% use personal dust sampling;
   (ii) Most facilities use gross alpha counting methods for assessing the alpha activity in dust samples;
   (iii) Most facilities use periodic monitoring for the assessment of exposure;
   (iv) Most facilities do not routinely use bioassay monitoring techniques, although some facilities are using urine analysis;
(d) Assessment of internal exposure from the inhalation of short-lived radon decay progeny:
   (i) The most widely used monitoring technique is active workplace monitoring of radon progeny in conjunction with the use of timesheets, followed by active monitoring of radon progeny using personal dosimeters;
   (ii) The most widely used monitoring strategy is work group averaging, followed by individual monitoring;
   (iii) Most facilities do not use background subtraction, which may lead to some overestimation of the dose;
(e) Dose assessment:
   (i) The most widely used method for the determination of occupancy time is the timesheet method, followed by the use electronic devices;
   (ii) While various dose conversion factors are being used, most facilities use factors specified by the regulatory body or by international recommendations or standards;

(iii) There is a need for global harmonization with respect to the selection of dose conversion factors in order to provide a common basis for comparison;

(iv) In order to obtain a more reliable estimate of the dose from inhalation of radionuclides in airborne dust, parameters such as particle size, solubility and radionuclide composition should be included in the dose calculation.

NEW IAEA SAFETY REPORT ON OCCUPATIONAL RADIATION PROTECTION IN THE URANIUM MINING AND PROCESSING INDUSTRY

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Abstract

Uranium exploration and production activities occur at locations throughout the world and utilize a range of different approaches and techniques. The radiation protection of workers, in a manner consistent with the graded approach to regulation, is a critical element of the regulatory control process, having a direct influence on the operation, and there is a strong need to ensure that appropriate monitoring and control mechanisms are in place. New radiation safety requirements have been established in the latest version of the IAEA International Basic Safety Standards, published in 2014. As part of the implementation of these new requirements to industrial activities involving NORM, the IAEA has developed a new safety report on the control of occupational exposure in the mining and processing of uranium ore. This safety report is designed to provide information on appropriate methodologies for ensuring the protection of workers and is aimed at regulatory bodies, operators and workers. The report covers the various commonly used uranium mining and processing operations and techniques. It describes the general radiation protection considerations relevant to this type of industrial activity; the general methodology applicable to monitoring, control and dose assessment; the relevant exposure pathways; and the radiation protection programme.

1. INTRODUCTION

The Fundamental Safety Principles (IAEA Safety Standards Series No. SF-1) [1], together with Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards (IAEA Safety Standards Series No. GSR Part 3) [2], set out the principles and basic requirements for radiation protection and safety applicable to all activities involving radiation exposure, including exposure to natural sources. Implementation of the requirements for occupational radiation protection brings its own set of challenges to regulatory bodies, operators and workers. Experience stretching back more than 40 years has been gained in applying international radiation safety standards to uranium production facilities worldwide and regulations in many uranium producing countries are among the most comprehensive and stringent. Nevertheless, there is still scope for further optimization of the protection of occupationally exposed workers, for instance through improved mechanisms for reducing occupational exposure, achievement of informed personal behaviour and application of the best engineering controls.

While many uranium mining companies generally take active steps to reduce radiation exposure wherever and whenever they can, and often voluntarily adopt the most recent international recommendations on dose limits and necessary occupational radiation protection requirements before they become part of the local regulations, consideration needs to be given to further optimizing the radiation protection of workers on an industry-wide and global basis. This is important as it supports the implementation of internationally consistent standards and approaches regarding the protection of workers.

Uranium is a naturally occurring element with an average concentration of 2.8 ppm in the Earth's crust. Traces of it occur almost everywhere. Its abundance is greater than that of gold, silver or mercury; it is similar to that of tin; and is slightly less than that of cobalt, lead or molybdenum. It is mined and concentrated in a manner similar to that used for many other metals. Over the last 60 years, uranium has become established as the world's principal nuclear fuel and an increase in the rate of prospecting, mining and processing of uranium ore is inevitable.

There are three main methods of extracting uranium from the ground: underground mining, open pit mining and in situ leaching (sometimes referred to as in situ recovery). Uranium ore from mines is crushed, ground and then leached to dissolve the uranium and separate it from the host rock. (In the case of in situ leaching operations, these steps are not necessary because the uranium is already in solution.) The dissolved uranium is then separated by ion exchange before being precipitated, dried and packed. The product uranium oxide concentrate is also referred to as yellowcake and mixed uranium oxides U_3O_8 and/or UO_4.

In addition, uranium can be recovered as a by-product from phosphate fertilizer production and from the mining of other minerals including copper and gold where the ores contain economically exploitable quantities of uranium. In such situations, the treatment process to recover uranium may be more complex.

With the current level of interest in nuclear power, there has been an increase in uranium exploration and also in the development of new uranium mining and processing facilities in many countries. World production in 2012 was 58 344 t of uranium [3]. This uranium production occurred across nearly 20 countries at approximately 50 different mining and processing facilities. As a consequence, the numbers of workers in uranium mining and processing may increase substantially within the next few years. During uranium mining and processing, workers may be exposed externally to gamma rays emitted from the ore, process materials, products and tailings. In addition, they may be internally exposed through the inhalation of dust containing long-lived radionuclides, radon and radon progeny and through ingestion, injection and absorption of contamination.

2. OBJECTIVE OF THE SAFETY REPORT

The objective of the safety report is to provide detailed information that will assist regulatory bodies and industry operators in implementing a graded approach to the protection of workers against exposures associated with uranium mining and processing. This information will also serve as the basis for creating a common understanding, based on common knowledge, between the various stakeholders — such as regulators, operators, workers and their representatives, and health, safety and environmental professionals — of the radiological aspects of the various processes involved and the ways in which these aspects can be addressed appropriately and effectively.

3. SCOPE OF THE REPORT

The safety report describes the various methods of production used by the uranium industry and provides practical information on the radiological risks to workers in the exploration, mining and processing of uranium, on exposure assessment, and on management of exposure based on the application of the appropriate standards and good working practices. This information has been compiled from published literature, from unpublished data provided by contributors to the report and from numerous experts with extensive experience, notably in the various sectors of the uranium mining and processing industry.

4. STRUCTURE

The report comprises six sections. Following the introductory section, Section 2 gives an overview of the uranium industry and the general radiation protection aspects of uranium mining and processing stages and techniques. Section 3 summarizes the radiation protection considerations that apply to the uranium mining and processing industry in general and application of the international standards in particular, including the basic radiation protection principles, the graded approach to regulation and specific aspects of radionuclides in the uranium decay series. Section 4 addresses the general methodology for control with the introduction of occupational health and safety considerations, the hierarchy of control, the radiation protection principles and exposure pathways. Section 5 deals with the requirements and dose assessment, with discussion on general dose considerations for different types of exposure pathways. Section 6 introduces the essentials of radiation protection programmes to adequately protect the workers, illustrating the process, design and operation, principal exposure pathways, control mechanisms and monitoring and dose assessment for different uranium mining and processing stages and techniques. The report is supplemented by appendices describing the findings of the International System on Uranium Mining Exposures (UMEX) survey, and the technical details of various exposure pathways.

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DOSE ASSESSMENT IN A PHOSPHORIC ACID FACILITY IN ZIMBABWE

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Abstract

In a radiological survey of a phosphoric acid plant, the radiation doses were consistently low, in the range 0.05–0.26 μSv/h. The analysis of several materials collected in the facilities, including phosphate rock, phosphogypsum and scale in pipes and tanks, revealed low activity concentrations of uranium and thorium series radionuclides. These radionuclide concentrations were consistent with the low ambient radiation doses measured in the facilities. It was concluded that the risk of occupational radiation exposure in these facilities is very low. Unlike phoshogypsum from other regions worldwide that contain elevated radionuclide concentrations, the phosphogypsum from Zimbabwe has low activity concentrations and can be easily used as a co-product in other applications.

1. INTRODUCTION

Several industries in Zimbabwe, including phosphate mining and phosphoric acid production, may be classified as NORM industrial activities. According to international studies and guidelines, such activities may give rise to enhanced levels of radiation in the workplace and surrounding environment [1, 2]. A preliminary radiological assessment was carried out at a phosphoric acid plant near Harare to identify the radionuclides present and the exposure of workers.

The phosphoric acid plant has been in operation since 1958, using raw material from phosphate rock mines located about 150 km from Harare. From 2010 onwards, owing to a decline of economic activity, the production of phosphoric acid diminished significantly. Nevertheless, considering that agriculture is a key strategic sector in Zimbabwe, there is great optimism that fertilizer production will recover at the plant. Currently, this plant continues to produce single superphosphate fertilizer (SSP), a blended mixture of sulphuric acid and phosphate rock, without separation of phosphogypsum (Fig.1).

In the phosphoric acid plant there are facilities for the production of sulphuric acid from pyrite and sulphur, facilities for the production of phosphoric acid from sulphuric acid and phosphate rock, warehouses for storage of phosphate fertilizer, and residue piles of pyrite and phosphogypsum nearby. Several thousand tonnes of phosphogypsum from past operations are stacked in the vicinity of the plant. Phosphogypsum from these legacy piles is currently sold as fertilizer as well (Fig. 1). In addition, the production of aluminum sulphate is carried out at the site using imported bauxite. In the facilities there are also administration and laboratory buildings, while the residential compound for factory workers and their families is located close to the phosphogypsum piles. Radiation dose measurements and radiological risk assessments have not been made at this facility before and so a preliminary survey was carried out.

2. MATERIALS AND METHODS

In collaboration with the phosphate company and the Radiation Protection Authority of Zimbabwe, a radiation survey of the area was carried out in July 2015 and samples of various materials were collected for radionuclide analysis in the laboratory. Measurements of ambient radiation dose rates were carried out with a gamma spectrometer Identifinder (FLIR) and portable radiation dosimeters FH-40 (Thermo Scientific). Measurements were generally carried out 1 m above the ground and in contact with containers, metal reactor vessels and metal pipes in the phosphoric acid plant. Samples of phosphate materials, including the phosphate rock raw material, SSP fertilizer, phosphogypsum, and scale in pipes and pumps used to transfer phosphoric acid were collected in identified plastic bags.

Analyses of samples were carried out in the laboratory by high resolution gamma spectrometry and by alpha spectrometry. For radionuclide quantification by gamma spectrometry, aliquots of about 50 g of the sample materials were packed in cylindrical sample containers and closed tight. After allowing 35 days for the establishment of radioactive equilibrium between radium, radon and its progeny, the samples were measured by gamma spectrometry using BeGe large volume detectors (Canberra) duly calibrated with customized standard radioactive sources from Eckert & Ziegler. Gamma spectra analysis and computations were made using Genie2000 software (Canberra, Meriden, CT, USA). For the determination of radionuclides by alpha spectrometry, isotopic tracers (^{232}U, ^{229}Th, ^{224}Ra, ^{209}Po, and 10 mg Pb^{2+}) were added to a weighed aliquot of about 1 g of the sample material for determination of the analytical chemical yield. These aliquots were fully dissolved in mineral acids using microwave digestion and radionuclides were separated by radiochemical procedures using ion exhange columns and electrodeposited onto stainless steel polished discs. Radionuclide measurements were made by alpha spectrometry using ion implanted silicon detectors (ORTEC EG&G). Analytical quality control of the results was ensured through periodic analysis of IAEA certified reference materials and participation in international radioanalytical intercomparison exercises, with good results [3, 4].

FIG. 1. Production of SSP fertilizer (left); loading phosphogypsum from legacy stacks for use in agriculture (right).

3. RESULTS AND DISCUSSION

The radiation survey of the area — including the area outside the administration buildings, warehouses, acid reactors and steamers — generally indicated ambient dose rates in the range 0.05–0.26 µSv/h. Measurements made at specific locations around large objects displayed the following dose rates:

- Phosphate rock pile outdoors: 0.12–0.17 µSv/h;
- Phosphogypsum pile in the factory yard: 0.08 µSv/h;
- SSP pile in the warehouse: 0.13–0.17 µSv/h;
- Decantation tank for phosphoric acid (empty): 0.20 µSv/h;
- Metal pipes and pump for phosphoric acid transfer: 0.20–0.22 µSv/h.

For comparison, the ambient radiation dose rates in open areas in Harare were found to be 0.10–0.12 µSv/h at a public garden and 0.17–0.22 µSv/h in the streets of office and residential areas. It was concluded, therefore, that the areas around the phosphoric acid plant did not show any evidence of significantly enhanced of dose rates.

The results of the various sample analyses are shown in Table 1. Samples of solid materials analysed by gamma spectrometry displayed low concentrations of all radionuclides. Radium-226 from the uranium series and ^{228}Ra from the thorium series were present, both in generally low concentrations, while ^{235}U, the parent radionuclide of the actinium series, was below the detection limit for the gamma spectrometry analytical procedure used. From analysis by alpha spectrometry, the phosphate rock contained 17 ± 1 Bq/kg of ^{238}U. In some materials, the naturally occurring radionuclide ^{40}K was present in concentrations higher than those of the uranium and thorium series radionuclides. Attempts to determine ^{137}Cs concentrations indicated levels below detection limits. Therefore, the measured dose rate was almost entirely due to the naturally occurring radionuclides of the uranium and thorium decay series and ^{40}K, as expected. Bauxite, imported for the production of aluminum sulphate in the chemical plant, also displayed low uranium ant thorium contents.

TABLE 1. RADIONUCLIDE CONCENTRATIONS IN SOLID MATERIAL SAMPLES

	Mean activity concentration and standard deviation (Bq/kg)					
	^{40}K	^{137}Cs	^{226}Ra	^{228}Ra	^{235}U	^{238}U
Phosphate rock (raw)	<34	<1.4	8 ± 2	26 ± 2	<10	
Phosphate rock (sieved)	<35	<1.6	12 ± 4	28 ± 4	<10	
SSP fertilizer (1 day old)	<48	<2.2	<5.8	14 ± 3	<11	
Encrustations (pipe scale)	86 ± 30	<1.6	13 ± 3	44 ± 4	<11	
Encrustations (phosphoric acid tanks)[a]			3130 ± 160		15 ± 2	303 ± 13
Phosphogypsum	<36	<1.4	<5.1	<2.8	<9.2	
Bauxite (imported)	279 ± 54	<3.1	19 ± 6	<6.1	<18	

[a] Measured using alpha spectrometry.

These results show that the phosphate rock used in the production of phosphoric acid and SSP fertilizers in Zimbabwe has low levels of natural radionuclides. Indeed, ^{226}Ra concentrations of 8 ± 2 Bq/kg in phosphate raw material and <5.1 Bq/kg in phosphogypsum are not very common in this industry and rank low in comparison with worldwide values [1, 2, 5]. Consistent with these low radionuclide concentrations, radiation doses in the facilities and by the phosphogypsum piles near the plant were at background levels. Therefore, although this has been a preliminary survey, it seems highly unlikely that exposure of workers and members of the public would exceed the annual dose limit of 1 mSv for non-radiation workers (i.e. members of the public) specified in the International Basic Safety Standards [6].

4. CONCLUSIONS

The radiation survey in the facilities of phosphoric acid production plant and in phosphate fertilizer warehouses indicated low ambient radiation dose rates, generally similar to the natural radiation background. Phosphate fertilizers produced with the low radioactivity phosphate rock available in the country contain low levels of radionuclides and their repeated application in agriculture is unlikely to increase the uranium and radium content of soils. Furthermore, the low radioactivity content of the phosphogypsum in the legacy piles allows the use of this by-product in other applications without the risk of increasing the radiation exposure of the population. In particular, the current use of this phosphogypsum as a fertilizer does not pose a risk of enhancing radioactivity in crops, especially of ^{226}Ra through root uptake of this radioelement [7]. Scale formed from phosphoric acid in tanks and pipe surfaces has much higher concentrations of radionuclides and should be managed as a NORM waste.

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Poster Presentation

MEASURING ACTIVITY CONCENTRATIONS USING A HIGH PURITY GERMANIUM DETECTOR: EXTENDED SOURCE EFFICIENCY CORRECTION AND OPTIMIZATION OF SAMPLE POSITION

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Abstract

An experiment using several calibrated gamma-ray sources located at different positions around a high purity germanium detector was conducted. The efficiency calibration curve for each position was obtained. More than 15 gamma-ray energies for each position point were used. The efficiency calibration curves facilitate the determination of radionuclide concentrations in an extended source such as NORM. These curves are useful for the measurement of a sample that is small enough to be considered as a point source, in order to obtain the position which produces the larger number of counts for the gamma-ray energy of interest. Preliminary results of the resulting efficiency curves and their interpretation are presented.

1. INTRODUCTION

The determination of radionuclide activity concentrations is a task of interest in all processes related to the handling of radioactive materials For instance, nuclear medicine makes use of radioactive elements both for diagnostic and therapy processes. As a consequence of such a procedure, residues of radioactive materials can be generated at different locations and materials associated with the medical centre. Those residues should be controlled by measuring the concentrations of radioactive materials. Likewise, radionuclides of natural origin are monitored by measuring their activity concentrations at different locations. The determination of radionuclide concentration is important because it allows us to determine radiation exposures in different regions and thus to estimate the doses received by the population. This is particularly relevant for material associated with industrial processes such as mining, which may fall within the definition of NORM and for which monitoring of radionuclide concentrations may therefore be mandatory.

Several methods are available for determining the concentrations of radionuclides within a sample. One of them is gamma-ray spectroscopy, a method for determining the concentrations of gamma emitting radionuclides. There are several gamma-ray spectroscopy techniques. One consists of a comparison of the radiation emitted by a reference material with the radiation emitted by the sample. Nevertheless, the use of such a technique implies the equivalence of the self-absorption of the radiation emitted by the reference material and the sample. This requires a reference material for each sample type. Moreover, in many instances, various reference materials are required for the determination of different radionuclides**.**

To avoid the use of reference materials, the measurement system can be calibrated in such a way that the radiation measured can be converted into activity concentration. To achieve success with this approach, several considerations must be taken into account. Two of the more challenging considerations are the self-absorption effect and the efficiency correction.

The self-absorption correction can be performed by calculations using the attenuation coefficient, which requires knowledge of the elemental composition of the sample. A workaround with the self-absorption effect is to measure a small piece of sample in order to be able to neglect the effect without modifying the result of the measurement significantly. Nevertheless, as the radionuclide concentration becomes smaller, the radiation emitted by the sample becomes more comparable with the background radiation, requiring a long measurement time. The point where the sample radiation becomes similar to the background radiation represents the detection limit and for a small sample such a limit is easily reached. By using a bigger sample the detection limit will increase. However, the use of the efficiency correction for a large extended source is not an easy task. Previous work has shown that the measurement of the efficiency of a high purity germanium (HPGe) detector at several spatial points will allow the necessary efficiency correction to be performed when determining the concentrations of radionuclides hosted by an extended source [1].

In this paper, the efficiency correction when using an HPGe detector is discussed. The methodology used to determine the efficiency of an HPGe detector is described and the results of the efficiency measurements are discussed.

2. METHODOLOGY

Two types of efficiency parameters are generally referred to in gamma spectrometry: the absolute efficiency and the intrinsic efficiency [2]. The absolute or total efficiency ε_{tot} in the full energy peak (FEP) is the number of photons registered in the photopeak divided by the number of photons emitted by the source. This quantity can be factored into two parts: the intrinsic efficiency and the geometrical efficiency. Using an isotropic radiation point source, the intrinsic efficiency is defined as

$$\varepsilon_{\text{int}} = \frac{Photons\ registered\ in\ the\ FEP}{Photons\ impinging\ on\ the\ \det ector} \tag{1}$$

ε_{int} depends on the interaction cross section of the incident radiation with the detector. The geometrical efficiency is the fraction of the radiation coming from the source that is geometrically intercepted by the detector. It depends entirely on the geometric configuration of the relative position of the detector and the source: $\varepsilon_{geo} = \Omega/4\pi$, where Ω is the solid angle subtended by the sensitive zone of the detector at the location of the source:

$$\Omega = \int_{dS} \frac{\hat{n}}{\rho^2}\, dS \tag{2}$$

Two cases are considered for the calculation of the geometrical efficiency with respect to the point source. First, the solid angle on the sensitive zone detector ($r \leq R$), where R is the radius of the Ge crystal, and second, the solid angle between the lateral face of the detector to source ($r > R$). In this work some measurements over the end cap of the detector were conducted. Fig. 1 describes the variables implied to solve the solid angle, the ℓ and h variables related to the spatial location.

The HPGe coaxial detector used has a relative efficiency 40% compared with the standard 3"×3" NaI detector [3]. Seven point gamma-ray emitter sources ^{133}Ba, ^{109}Cd, ^{57}Co, ^{137}Cs, ^{60}Co, ^{22}Na and ^{152,154,155}Eu [4] were utilized, with the FEP between 53 and 1596 keV. The activity uncertainties were 20% for the ^{137}Cs source and 5% for the others.

Each of the seven calibration sources was placed on top of the detector at $h_1 = 45.35$ mm and $h_2 = 163.95$ mm, counted over a period of between 1 min to 3 h, depending on the source activity, at two horizontal locations $\ell_1 = 0$ and $\ell_2 = 20$ mm. The experimental setup is shown in Fig. 2, from which it can be seen that extended sources were used. The spatial distribution of the active material in the source was taken into account in the solid angle calculations.

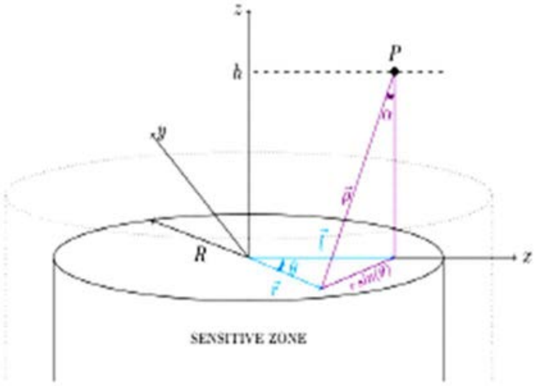

FIG. 1. Solid angle description.

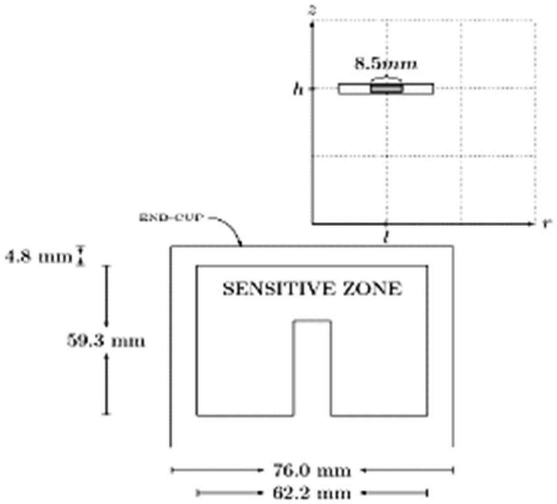

FIG. 2. Experimental setup (from Germanium Detectors, User Manual, Canberra Industries, 2003*).*

3. RESULTS

The resulting efficiencies as a function of the energy are shown in Figs 3 and 4. These are the intrinsic efficiencies and therefore they are already corrected for solid angle. From Fig. 3 it can be seen that, for energies greater than approximately 200 keV, the efficiency increases slightly when the position of the source changes from ℓ_1 to ℓ_2. Because of the size of the uncertainties, Fig. 4 does not allow certain conclusions to be drawn.

The same efficiency curves were used to compare two different fixed heights with different horizontal positions. First, we compare two different heights, for the same value of ℓ. The measured efficiency is larger for h_2 than for h_1. Fig. 4 shows slight variations when the horizontal position of the source is changed at a fixed height. Reference to Fig. 5 provides a better understanding of Figs 3 and 4. For ℓ fixed at two different heights, the shortest path for a gamma ray traversing the crystal occurs at lower heights. It implies that such a gamma ray can travel through longer paths to greater heights, therefore it has a greater probability of being detected: $P1<P2$. On the other hand, by varying ℓ at fixed h there will be a different value for the largest distance that a gamma ray can travel inside the crystal. This is illustrated in Fig. 5 with the relation $D1<D2$. This implies that most gamma rays will have a greater probability of being detected and this effect will more evident for higher energy gamma rays. To obtain more conclusive data about the last analysis it would be necessary to carry out measurements for greater values of ℓ. Spatial variations in the measurement efficiency are expected to be used to calculate the concentrations of radionuclides of natural origin in extensive sources as was shown in Ref. [1].

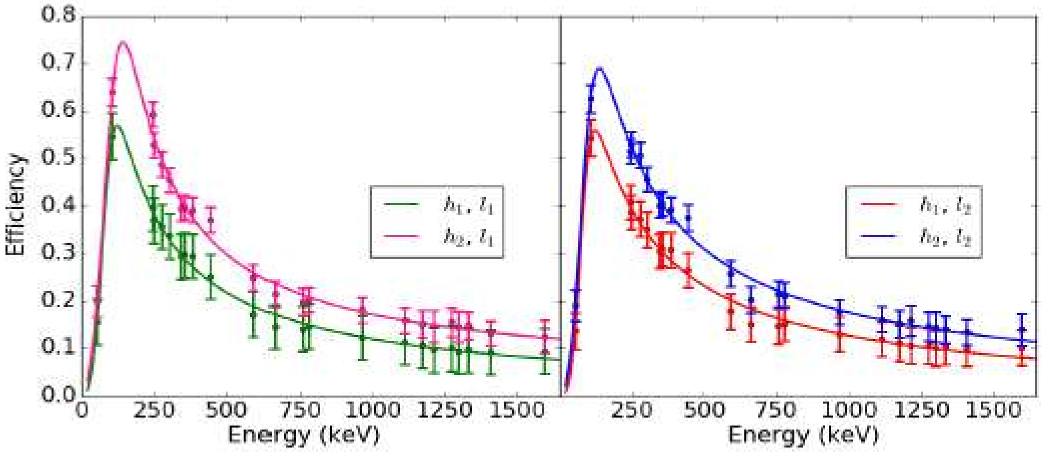

FIG. 3. Comparison of intrinsic efficiency at different heights.

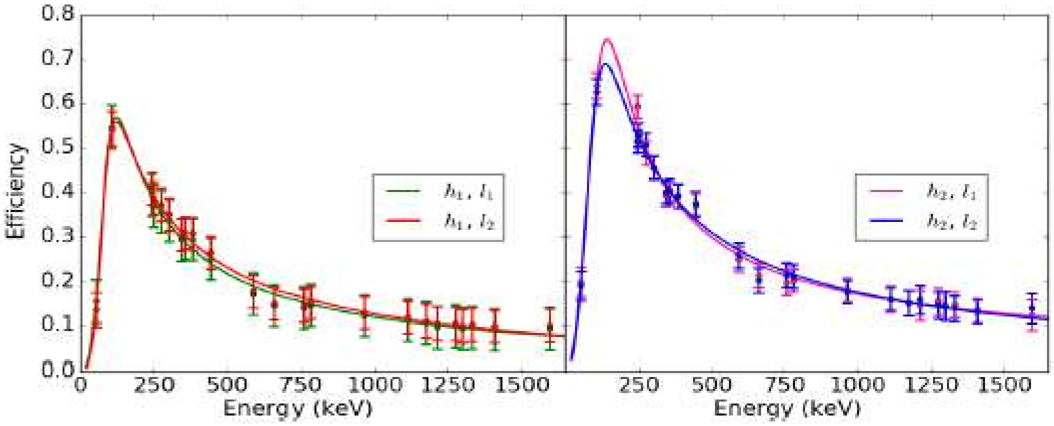

FIG. 4. Comparison of intrinsic efficiency at different axial positions.

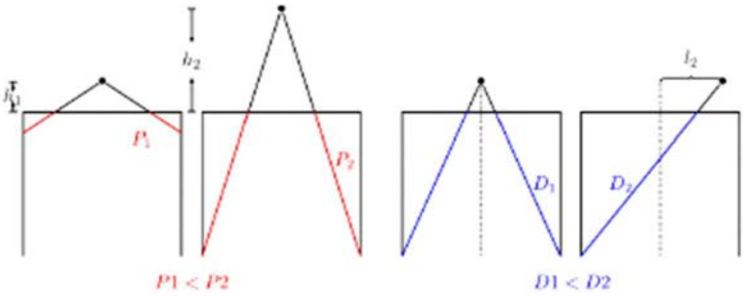

FIG. 5. Schematic gamma ray paths for different source positions.

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Poster Presentation

DETERMINATION OF THE MINIMUM SAMPLE MASS OF U_3Si_2 TO BE USED AS CANDIDATE REFERENCE MATERIAL FOR CHEMICAL ANALYSES OF TOTAL URANIUM AND TOTAL SILICIDE

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Abstract

To guarantee the reliability and comparability of analytical data, the use of reference materials is essential. Uranium intermetallic compounds, in particular U_3Si_2, have been the preferred fuel materials for high flux research reactors. A method for the quantitative determination of the minimum representative sample mass of a candidate for reference material for determination of total uranium and silicide is described and illustrated in this paper. The analytical method used for determination of total uranium was the high precision potentiometric titration method of Davies and Gray, while total silicide was determined gravimetrically. The study was conducted according to ISO Guide 35. A preliminary test for homogeneity can be performed after homogenization as an integral part of the candidate reference material preparation.

1. INTRODUCTION

Uranium intermetallic compounds, in particular U_3Si and U_3Si_2, are the preferred choice of fuel materials for high flux research reactors. Uranium silicide fuel has been conventionally prepared by rolling or extruding the blended powders of uranium silicide and aluminum. The use of reference materials (RMs) and, where possible, certified reference materials (CRMs) enable a laboratory to provide the results of analytical measurements with an acceptable level of reliability [1, 2]. The preparation of these materials should consider not only all certification parameters (values and corresponding uncertainties) but also other information on the handling and use of a CRM. An extremely important part of this information is the 'minimum sample size' of a solid CRM [1–5]. Quantitative statements based on micro- or macro-homogeneity determinations are rarely made. As a direct result, most CRMs should not be used to calibrate or control micro-techniques. It is impossible to assess the uncertainty to be assigned to the certified value if samples smaller than the minimum sample size are used [4, 5]. Thus, this study was conducted according to ISO Guide 35 as a preliminary test to elucidate the minimum sample size to be used for a repeatable candidate reference material [3, 4, 6].

2. METHODS

2.1. Minimum sample mass

According to ISO Guide 35 [3] there are two ways to realize a minimum mass assay. A preliminary test for homogeneity can be performed after homogenization as an integral part of the CRM preparation process. Alternatively, by taking a vial to be used in the analysis, a 'sub-sample' bottle with a different mass is made. As the size of this sub-sample is decreased, it can be determined whether this subdivision into progressively smaller sub-samples results in variations in the uncertainty mentioned in the certificate (typically for a 95% confidence interval), giving instead a range of statistical tolerances of 95% [3–6]. The size of the sample for a minimum amount of CRM corresponds to a sample mass *m* for which the uncertainty expressed in Eq. (1) becomes equal to that of Eq. (2).

$$UNC = \frac{\pm t_{1-\alpha} s}{\sqrt{n}} \tag{1}$$

where t is the Student t-factor at a probability level of $1-\alpha$, s is the experimental standard deviation and n is the number of measurements for certification, assuming for simplicity that n measurements are performed under the same conditions by an impartial method [4], and

$$\Delta = k_2' s \tag{2}$$

where k_2' is a factor for the tolerance limits of normal distributions for both sides, with at least a proportion p for a probability level of $1-\alpha$ and a series of n samples analysed during the study uniformity, s is the experimental standard deviation for the study homogeneity [4]. The second method is conducted by selecting an experimental mass range based on the analytical method used in the validation process.

2.2. Determination of total uranium by the high precision potentiometric titration method of Davies and Gray

The uranium titration method introduced by Davies and Gray [7] (and subsequently improved [8]) is the most widely used analytical method for the potentiometric titration of uranium from nuclear materials. It relies on the reduction of U (VI) to U (IV) followed by a subsequent titration of U (IV) with potassium dichromate. The result is given by the following equation:

$$\%U = 100 \times \frac{mg\ U\ found}{(1000) \times (solution\ aliquant\ wt,\ g) \times (DF)} \tag{3}$$

where *DF* is the dilution factor expressed as:

$$DF = \frac{g\ sample\ taken}{total\ g\ sample\ solution\ prepared} \tag{4}$$

2.3. Gravimetric determination of silicide

The total silicide is determined gravimetrically by an indirect method, dissolving an aliquot of the solid sample U_3Si_2, hydrofluoridization, and then insolubilization of silica. The result is obtained by measuring the mass of Si containing impurities ($m1$); volatilization of silicon tetrafluoride, measurement of the mass $m2$ of the residue containing impurities and finally the calculation of the percentage of pure Si in the sample, considering the difference between $m1$ and $m2$, and the mass ma of the solid sample [9]:

$$\%Si = \frac{(mi - mf) \times f_{Si} \times 100}{ma} \tag{5}$$

3. RESULTS AND DISCUSSION

Bottle 11 was used for the study of homogeneity within the bottle. Three mass sub-samples containing 0.5, 0.8 and 1.2 g of U_3Si_2 were used for total uranium and silicide analysis. Dixon tests were used for evaluation of the results, Shapiro-Wilks for normal 95% confidence, and ANOVA for equality between the average percentages of the elements; F calculated was lower than the critical F for 95% confidence [4, 5]. There was no significant difference between the mean values in the variation of the mass of uranium elements and total silicide (see Table 1).

TABLE 1. PERCENTAGE OF TOTAL URANIUM MASS OBTAINED BY THE DAVIES AND GRAY METHOD AND TOTAL SILICON OBTAINED BY THE GRAVIMETRIC TECHNIQUE

	Total uranium (%)			Total silicide (%)		
	1.2 g	0.8 g	0.5 g	1.2 g	0.8 g	0.5 g
Average ± SD	91.84 ± 0.01	91.69 ± 0.02	91.58 ± 0.11	7.62 ± 0.02	7.63 ± 0.03	7.62 ± 0.01
RD	0.01	0.02	0.12	0.20	0.41	0.19
W	0.62	0.71	0.86	0.90	0.75	0.92
P-valor	0.0003	0.0005	0.11	0.45	0.012	0.46
Dcalc	0.12	0.16	0.06	0.18	0.06	0.3
Fcalc[a]	1.10	0.004	1.18	0.97	0.96	0.97
U (%)[b]	0.2	0.3	0.4	0.3	0.3	0.3

[a] Fcrit U total = 3.03; Fcrit Si = 5.41.

[b] *U(%)* – uncertainty.

The ideal minimum mass was estimated by comparison, in percentage form, DPR with the dispersion of individual RSD values and mean for each selected mass in repeatability conditions, with 8 replicates of titration and weighed at the same time and by the same analyst. A graphical comparison of the results for selected elements is shown in Fig. 1. It was observed that, for total uranium, the deviation ranged from 0.05 to 0.28%, with an average value of 0.16 ± 0.04%. For total silicide, the deviation ranged from 0.08 to 0.84%, with an average value of 0.35 ± 0.7%. Differences in the average values of the elements for the masses of 0.5, 0.8 and 1.2 g were not statistically significant. However, the individual results for the standard deviation were more widely dispersed. Thus 1.2 g was selected as the minimum mass of the sample for characterization of the material, since it resulted in individual values that were mutually compatible.

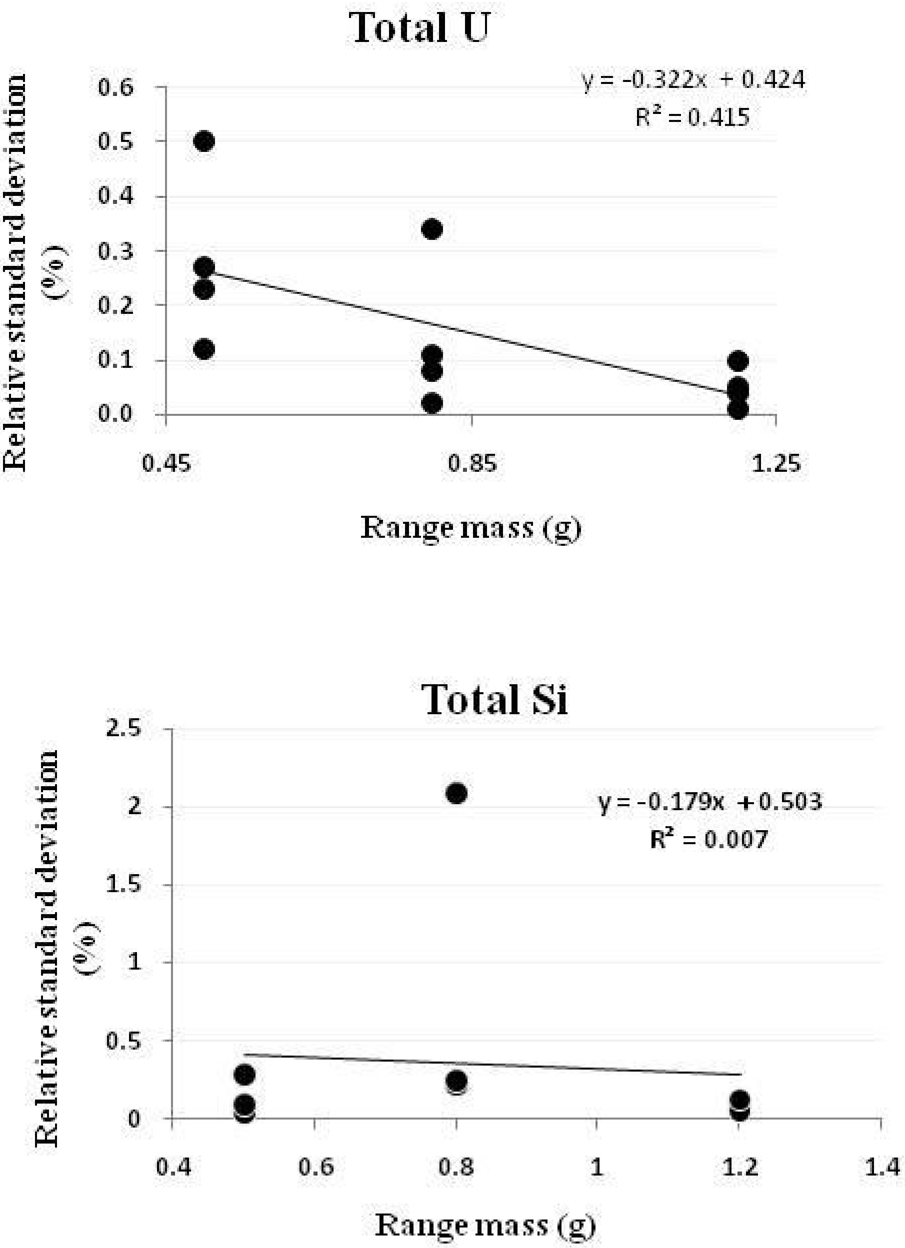

FIG. 1. Standard deviation in relative weight function for the study of homogeneity in the vial using Davies and Gray test for total uranium and gravimetric assay for total silicide.

ACKNOWLEDGEMENTS

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Poster Presentation

SELF-ATTENUATION FACTORS IN GAMMA-RAY SPECTROMETRY OF SAMPLES CONTAINING RADIONUCLIDES OF NATURAL ORIGIN

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Abstract

High resolution gamma-ray spectrometry is currently the most widely used analytical technique for qualitative and quantitative determination of radionuclides. Quantification of elements relies on the correct analysis of the spectra, depending strongly on the efficiency calibration of the measurement apparatus, most often performed with aqueous standard multi-radionuclide solutions. For efficiency calibration curves obtained by this method and for samples containing radionuclides of natural origin such as sand, soil, rocks and wall paint with apparent typical densities higher than that of water, self-attenuation correction factors were experimentally determined for hundreds of different samples using the Cutshall transmission technique. The results show that, to obtain more reliable analyses, correction factors for the self-attenuation behaviour, especially in the lower part of the energy spectrum, should be used. Since attenuation depends not only on the density of the sample but also on its chemical composition, correction factors have to be determined for each sample.

1. INTRODUCTION

It is well known that natural radioactivity is ubiquitous in the environment, mainly in minerals such as sand, soil and rocks. High resolution gamma-ray spectrometry is currently the most widely used analytical technique for qualitative and quantitative determination of natural radioactivity in such materials [1, 2]. Quantification of elements relies on the correct analysis of the gamma spectra and depends strongly on the efficiency calibration of the measurement apparatus, which is usually performed with aqueous standard multi-radionuclide solutions.

Owing to self-absorption within the sample, lower energy gamma rays have less penetrating ability and tend to interact more readily with matter. So, when an efficiency calibration curve is obtained with an aqueous standard multi-radionuclide solution, a self-attenuation correction is required if samples present densities higher than those of water. Environmental samples such as sand, soil and rocks [3–5] or samples of manufactured products such as wall paint [6] containing radionuclides of natural origin usually have apparent densities higher than water, so correction of the efficiency curve is required for accurate characterization of the material [7, 8]. The full-energy peak efficiency of low energy emitters in semiconductor gamma spectrometers depends strongly on a number of factors including sample composition, density, sample size and gamma-ray energy. Several methods can be used for accurate determination of self-attenuation in the sample, such as computer simulation, the use of spiked or natural matrix reference materials that match each sample type to be analysed, the use of sets of gamma absorption curves, or direct gamma-ray transmission measurements for each sample [9–11].

The environmental laboratory at Instituto de Pesquisas Energéticas e Nucleares (IPEN) is currently using the transmission technique for its low cost and practical application [3–6, 12, 13]. Self-attenuation correction factors for samples of sand, soil, rocks and wall paint have been determined experimentally using high resolution gamma-ray spectrometry. For all samples with apparent densities higher than 1 g/cm^3, correction factors have to be determined for each sample since attenuation strongly depends not only on the density of the sample but also on its chemical composition.

2. GAMMA-RAY TRANSMISSION METHOD

The Cutshall technique [11] basically consists of measuring the transmission of gamma-rays through both the sample of interest and an ultra-pure water sample of the same geometry [12, 13]. IAEA gamma-ray sources of ^{152}Eu, ^{60}Co and ^{137}Cs, with energy peaks ranging from 122 keV to 1408 keV, are generally used in order to cover the range of energies of the radionuclides of interest. However, if assessment of only ^{210}Pb is required, only a ^{210}Pb source is needed. For each sample and specific density, a self-attenuation factor was obtained for each gamma transition energy of the sources, using Eq. (1).

$$f_i = \frac{\ln\left(S_i/W_i\right)}{\left(S_i/W_i - 1\right)} \tag{1}$$

where f_i is the self-attenuation factor for the i^{th} gamma transition, S_i is the beam intensity transmitted through the sample for the i^{th} gamma transition and W_i is the beam intensity transmitted through the ultra-pure water sample for the i^{th} gamma transition. A self-attenuation function was fitted for each sample, considering the attenuation factors acquired for all gamma transitions.

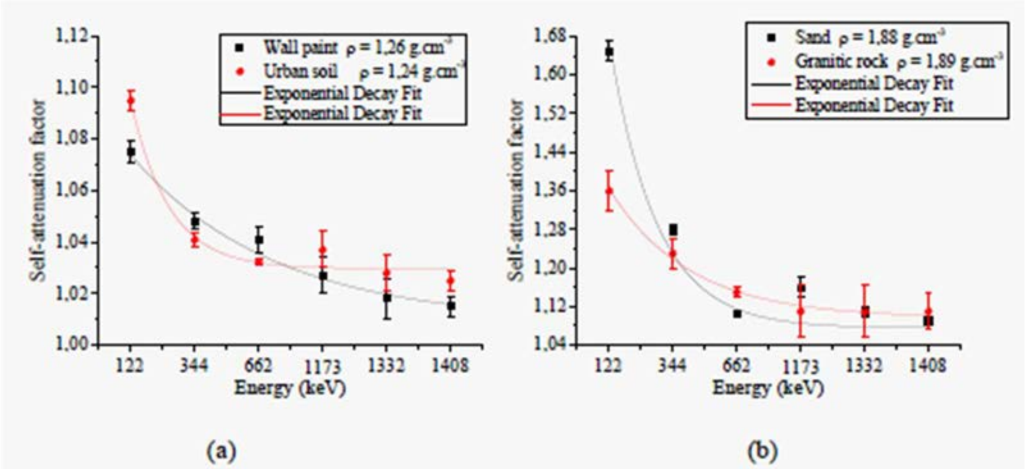
3. RESULTS AND DISCUSSION

The samples analysed over the last seven years at the Environmental Radiometric Laboratory of IPEN are summarized in Table 1.

TABLE 1. TYPES OF SAMPLES ANALYSED AND THEIR DENSITIES

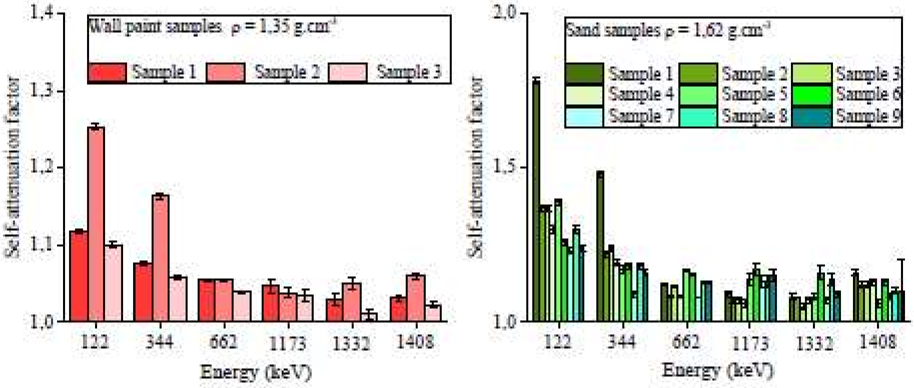
	Density (g/cm3)	Total number of samples
Granitic rock	1.57–2.02	50
Sand	1.26–2.35	132
Urban soil	1.07–1.32	10
Wall paint	0.97–1.46	50
Soil and sedimemt	1.3–1.8	165

Figure 1 shows typical fitted self-attenuation curves in the range 122–1408 keV for wall paint, urban soil, sand and granitic rock samples with similar densities. As expected, the attenuation factors are higher for lower energies but depend also on the sample type. For energies in this range, the strong dependence on the sample chemical composition (in addition to energy) is shown in Fig. 2 for samples of wall paint and sand of the same density. Self-attenuation factors for the 46 keV gamma transition of ^{210}Pb for soil and sediment samples of various densities are shown in Fig. 3.



(a) (b)

FIG. 1. Self-attenuation curves: (a) wall paint and urban soil, (b) sand and granitic rock.



(a) (b)

FIG. 2. Self-attenuation factors for samples of similar density: (a) wall paint, (b) sand.

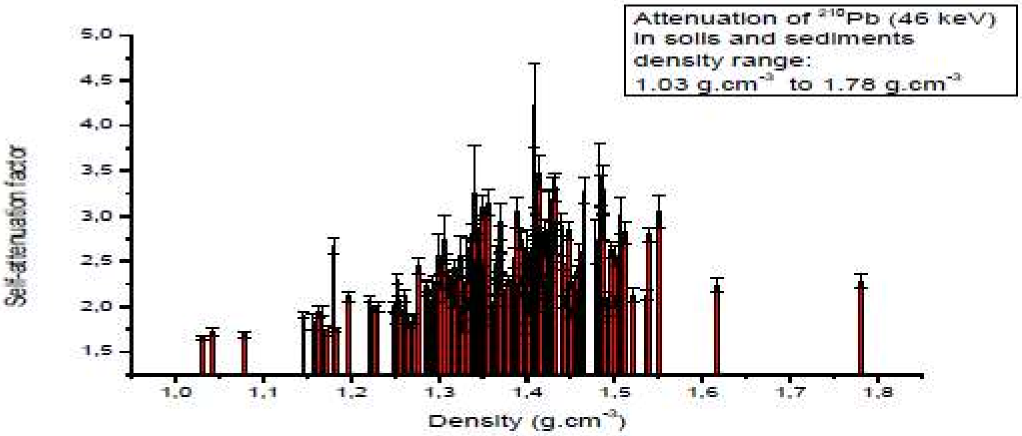


FIG. 3. Self-attenuation factors of ^{210}Pb for sediment and soil samples of different densities.

4. CONCLUSIONS

The self-attenuation correction factors determined by high resolution gamma-ray spectrometry for samples with high apparent densities such as sand, soil, rock and wall paint confirm that attenuation strongly depends on not only the density but also the sample chemical composition, so correction factors have to be determined for each sample. Such correction factors are needed not only for samples containing radionuclides of natural origin but for all samples with high densities when the efficiency curve is obtained with aqueous standard multi-radionuclide solutions.

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DECOMMISSIONING AND REMEDIATION

(Session 8)

Chairperson

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DECOMMISSIONING OF A RARE EARTHS EXTRACTION PLANT AND REMEDIATION OF THE NORM CONTAMINATED SITE

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Abstract

This paper describes the decommissioning of a monazite cracking plant for the extraction of rare earth elements, which had been in operation from 1982 to 1994. The decommissioning process was carried out in two phases: (i) rehabilitation of the plant site, and (ii) disposal of the thorium hydroxide (NORM) waste into a near-surface disposal facility. The decommissioning, decontamination and disposal activities commenced in September 2003. The activities carried out during the first phase included demolition of the plant building, excavation of contaminated soil, backfilling the site with normal soil and disposal of the waste. The aim of the site remediation was to reduce the radiation levels to normal background levels. Radiation levels were measured by means of an external gamma survey. On completion of the remediation, the result was verified by the regulatory body and the site was confirmed to be free of contamination and released from regulatory control. Monitoring of the site is being continued for another two years.

1. INTRODUCTION

The Malaysian minerals industry involves the import or local mining of raw material, transport of the material to a process plant, storage of raw material, and processing of the raw material to produce products and process residues. Large amounts of residues arising from the operation and decommissioning of mines and mineral processing plants may contain elevated levels of radionuclides of natural origin and, depending on the activity concentrations, may fall within the definition of NORM. Such residues exist mainly in the form of scale and sludge from the oil and gas industry, thorium hydroxide from the processing of xenotime and monazite, and iron oxide and red gypsum from the processing of ilmenite. Other types of residue include tin slag produced from the smelting of tin, as well as ilmenite, zircon, and monazite resulting from the processing of tin tailings (‘amang’). Some of these residues may be suitable for recycling to other industries or for direct use as by-products. If there is no prospect for recycling or by-product use, they are designated as waste and have to be disposed of accordingly. Environmental issues pertaining to waste disposal are of major concern to members of the public. Consequently, the management of NORM waste is the main issue associated with mining and mineral processing activities.

Currently, one mineral processing plant generating NORM waste from rare earths extraction has been decommissioned. A facility exists for disposal of the NORM waste resulting from these decommissioning and decontamination activities. This facility contains two engineered cells, the first of which contains contaminated soil and construction material from the decommissioning activities and the second of which contains thorium hydroxide waste generated during the operation of the plant.

2. LEGAL REQUIREMENTS FOR NORM RESIDUES

The Atomic Energy Licensing Act (Act 304 of 1984) is the main legal instrument for providing for the regulation and control of atomic energy, for the establishment of standards on liability for nuclear damage and for matters connected therewith or related thereto. The Atomic Energy Licensing Board (AELB) was established under Section 3 of the Act on 1 February

1985 to enforce the requirements of this legislation. It has adopted and enforced the following regulations for this purpose:

(a) Radiation Protection (Licensing) Regulations 1986,
(b) Radiation Protection (Transport) Regulations 1989,
(c) Atomic Energy Licensing (Basic Safety Radiation Protection) Regulations 2010,
(d) Atomic Energy Licensing (Radioactive Waste Management) Regulations 2011.

In terms of Act 304, the AELB regulates the various NORM activities — including oil and gas extraction, the processing of minerals containing radionuclides of natural origin, and the management of NORM residues — through licensing procedures and conditions. The licensing process covers the successive stages of facility siting and construction, facility operation, storage of process materials and, finally, decommissioning and decontamination (D&D) activities including the disposal of any NORM waste.

Radioactive waste management is governed by the Atomic Energy Licensing (Radioactive Waste Management) Regulations 2011. In terms of the second schedule of these regulations (and in accordance with international standards [1]), material with radionuclide activities or activity concentrations below the clearance levels of 1 Bq/g for individual radionuclides in the uranium and thorium decay series and 10 Bq/g for ^{40}K may be released from regulatory control. Thus, in order to be free from regulatory control, residues that have been recycled for use in other industries or are used directly as by-products must meet the clearance criteria — this may necessitate a reduction in activity concentration by suitable treatment.

If the clearance level is exceeded, the material is regarded as NORM and its processing or disposal is subject to regulatory control by AELB through the licensing process. For the disposal of NORM waste, the following options are available:

(a) For oil and gas sludge with moderate activity concentrations, disposal at a landfill site;
(b) For oil and gas sludge with higher activity concentrations, incineration, followed by disposal of the ash (in which the radionuclides have become concentrated) at a secured landfill site;
(c) For NORM waste from monazite cracking, a near-surface disposal facility.

3. DECOMMISSIONING AND DECONTAMINATION OF THE RARE EARTHS EXTRACTION FACILITY

A monazite cracking plant to extract rare earth elements commenced operation in the northern part of Malaysia in 1982. The company ceased operation in January 1994. Waste in the form of thorium hydroxide sludge (containing thorium and radium) was generated during the life of the facility, and various materials and equipment within the plant area became contaminated with thorium.

The D&D process started in September 2003. The sludge and contaminated material were temporarily stored in 85 000 drums (200 L capacity) at a ‘long term storage facility’ (LTSF) located about 10 km from the plant site, pending transfer to a near-surface disposal facility. The aim of the first phase of the D&D project (‘Plant D&D’) was to return the plant site to its original radiological condition and to dispose of the contaminated material in a part of the near-surface disposal facility designated as ‘Engineered Cell 1’ (EC 1). The material disposed of consisted mainly of contaminated construction material from the plant and contaminated soil from the site. The activities carried out included the demolition of the plant building, excavation of contaminated soil, backfilling the excavated area with normal soil, transporting the waste, and disposal of the waste into the disposal facility. This first phase was completed in October 2005.

The second phase of the project ('LTSF D&D') was to transfer and dispose of the thorium hydroxide waste (temporarily stored in the LTSF) into a part of the disposal facility designated as 'Engineered Cell 2' (EC 2). This waste was classified as low level radioactive waste with long-lived radionuclides of the ^{238}U and ^{232}Th decay series. The second phase started in 2011 and was completed at the end of 2014. The activities carried out were the decommissioning of the LTSF, waste categorization, waste stabilization (conditioning) and packaging of the waste in concrete containers prior to placement in the disposal cell. Finally, a single cap was to be installed to cover both EC 1 and EC 2.

4. DISPOSAL FACILITY (ENGINEERED CELL)

The near-surface disposal facility was constructed at the same site as that of the LTSF. The safety assessment, design and construction were carried out by a company based in the United States of America. The capacities of EC 1 and EC 2 are about 85 000 m^3 and 55 000 m^3, respectively. EC 1 was constructed adjacent to the LTSF site and EC 2 was constructed at the LTSF site itself. The inventory of waste is shown in Table 1.

TABLE 1. WASTE INVENTORY OF THE NEAR-SURFACE DISPOSAL FACILITY

	Waste	Volume (m^3)
Phase 1: Plant D&D	Contaminated soil excavated	66 000
	Contaminated concrete and rubble	6330
	Other contaminated material	2000
Phase 2: LTSF D&D	Thorium waste	16 200
	Contaminated material	10 000
	Contaminated rubble and other material	20 200

5. REMEDIATION OF THE PLANT SITE

The radiation levels at the plant site were measured by means of an external exposure survey. The site was divided into a 100×100 m grid and the external radiation levels were measured in each grid square. The contaminated soil was excavated and the area backfilled with normal (uncontaminated) soil. Post-remediation monitoring was to be continued for a further two years. The monitoring results were verified by the regulatory body as being below the applicable limit (see Table 2). The site was confirmed free of contamination and was released from regulatory control.

TABLE 2. SITE MONITORING RESULTS ON COMPLETION OF REMEDIATION

	Monitoring result	Limit	Basis for limit
Annual dose from external radiation (mSv/a)	0.4–0.5	1.0	Act 304
Ra-226 activity concentration in soil (Bq/kg)	90	100	Normal soil in Malaysia [2]

The licensee implemented a monthly programme of post-remediation monitoring (see Table 3) approved by the regulator and supervised by a consultant. The regulator also conducted independent site monitoring to verify the results.

TABLE 3. MONTHLY POST-REMEDIATION MONITORING PROGRAMME

	Parameter	Method
External radiation	Dose rate	TLD
Soil	Activity concentration, Ra-226 and Ra-228	Gamma spectrometer
Water	Activity concentration, Ra-226 and Ra-228	Liquid scintillator
Air	Activity concentration, Rn-222 and Rn-220 progeny	Radon–thoron detector

6. REGULATORY CONTROL

On-site routine inspection was carried out by the regulatory body to ensure the safety of the D&D activities in terms of radiation protection. The activities inspected were soil excavation, transport of the waste to the disposal facility, loading and unloading of the waste into and out of the transport vehicle, backfilling of the excavated area, construction of the engineered cell and placement of the final cap over it. Regular auditing by a consultant was also carried out to ensure compliance with all regulatory requirements.

7. CONCLUSION

Post-closure monitoring of the disposal facility is to continue for two years beyond its closure. Samples of flora, fauna, soil and water are collected and analysed to detect signs of radionuclide migration as a result of any leakage from the disposal facility. All radiation monitoring reports are submitted to the regulatory body at regular intervals as agreed by both parties. Institutional control over the closed disposal facility will continue for at least 300 years.

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EVALUATION AND SURVEY OF NORM LEGACY SITES IN AUSTRIA
(Summary)

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At the end of the 19th century several factories and institutes in Austria started to work with materials containing elevated concentrations of radionuclides of natural origin (especially isotopes of radium and thorium). In accordance with the European Council Directive 2003/122/Euratom on the control of high activity sealed radioactive sources and orphan sources, several campaigns to identify potentially contaminated sites in Austria were performed and the results were listed in a catalogue for (possible) radiologically relevant legacy sites. Based on the legacy catalogue, a study concerning the premises of a former chemical factory was carried out by the Austrian Agency for Health and Food Safety (AGES). According to historical research the company (founded around 1890) used to process pitchblende (uranium ore) residues for the production of radium as well as monazite sand for the production of thorium and subsequent manufacturing of incandescent gas mantles. The results of the radiological survey showed elevated concentrations of radionuclides of natural origin, especially isotopes of thorium, uranium and their progeny. In order to evaluate further sites of the legacy catalogue in a systematic way, a standardized screening procedure for NORM legacy sites has been developed and implemented.

At first the legacy site has to be identified — mostly through research. In Austria this is carried out in cooperation with the Austrian Federal Ministry for Agriculture, Forestry, Environment and Water Management. Based on the results, a legacy catalogue was developed and is periodically updated. The legacy sites are prioritized based on the radiological risks to the population. The dose limit for individual members of the public is 1 mSv per year. The guidance level for natural sources of radiation in Austria is 1 Bq/g. If the activity concentration is lower than 1 Bq/g for all radionuclides it can be assumed that the dose received by members of the public is lower than 1 mSv per year. If the activity concentration is higher than 1 Bq/g, a detailed exposure scenario has to be developed.

The next step is the characterization of the legacy site and the development of a sophisticated exposure scenario (e.g. through simulations with the computer code ResRad). The radiological characterization of a contaminated is often carried out in collaboration with companies specialized in environmental surveys of chemically contaminated legacy sites to evaluate a potential correlation between chemical and radiological contamination and therefore to determine all types of risk that could affect the population.

Based on the results of the characterization, a decision has to be made whether to decontaminate or secure the legacy site. For this purpose several factors have to be considered. For instance: Is the contamination indoors or outdoors? What is the size of the affected area? Is the material soluble or insoluble? Could the groundwater be affected by the contaminated material?

During the remediation of a contaminated site the following steps have to be taken:

(a) A risk based dose assessment for all involved parties has to be carried out;
(b) During the remediation work, radiation protection has to be ensured for all parties and the contamination status has to be monitored at all times (e.g. through continuous wipe tests);
(c) The cooperation with decontamination experts and other involved companies has to be coordinated;
(d) The success of the remediation has to be confirmed through comprehensive sampling of environmental media such as soil, water and air;
(e) Information on potential health risks has to be provided to the exposed population;
(f) The accumulated waste has to be disposed of in a proper way.

Throughout the screening process all the identified contaminated material has to be disposed of in a landfill suitable for NORM or classified as radioactive waste. In Austria these decisions are made on a case by case basis after a dose assessment. An adopted NORM waste strategy (according to Art. 102) on existing exposure will be developed within the implementation process of Council Directive 2003/122/Euratom.

HYDROGEOCHEMICAL AND ISOTOPIC STUDY OF GROUNDWATER IN THE ENVIRONMENTAL REMEDIATION OF OSAMU UTSUMI URANIUM MINE

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Abstract

The Osamu Utsumi mine, belonging to Brazilian Nuclear Industries, was in operation between 1982 and 1995, when uranium mining and processing ceased. During the stripping operations before mining, about 30 million m^3 of waste rock were removed. The waste rock dumps were deposited in valleys adjacent to the open pit. Since its shutdown, the mine has been going through an active maintenance system, the main focus of which is the treatment of acid mine drainage generated in the open pit and the waste rock dumps. Knowledge of the hydrogeology and the reactions involved in the generation of acid mine drainage is essential in the development of solutions aimed at minimizing the environmental impact and costs associated with the liabilities. The PHREEQE program allows the geochemical processes that occur between water and rock to be simulated, to determine the speciation of aqueous components and to calculate minerals saturation indices.

1. INTRODUCTION

The Osamu Utsumi Mine is in an area called Campo do Cercado located on the Planalto de Poços de Caldas, in the south-east of the state of Minas Gerais. The mine is part of the mining and processing complex of Poços de Caldas owned by Brazilian Nuclear Industries (INB). In 1977, stripping operations and activities of the processing complex were initiated and the complex was officially inaugurated in 1982, being exploited until 1995. It is estimated that during the operation of the mine, 94.5 million t of rock were removed, 2% of which was sent for processing and the rest deposited in piles of non-mineralized and mineralized rock. The non-mineralized rock was deposited in the valleys adjacent to the open pit, transforming these valleys into large rock repositories. The main waste rock piles from the point of view of environmental protection are waste pile 4, occupying an area of 56.9 ha and containing 12.4 million m^3 of non-mineralized rock with a slope height of 90 m and an inclination of 70°, and waste pile 8, occupying an area of 64.4 ha and containing 15.0 million m^3 of non-mineralized rock [1]. Since cessation of operations, the mine has been going through an active maintenance system, the main focus of which is the treatment of acid mine drainage (AMD) generated in the open pit and the waste rock piles. Knowledge of the hydrogeology and the chemical reactions involved in the generation of AMD is essential in the development of solutions aimed at minimizing the environmental impact and costs associated with the liabilities. Thus the main purpose of this study is to provide information that will enable the hydrochemical characteristics of the groundwater to be understood.

2. METHODS

The mining and processing complex occupies an area of approximately 15 km². The area that was the subject of this study (the open pit mine and waste pile 4) occupies an area of approximately 2.5 km² on the watershed of two major hydrographic basins: the Antas Stream basin and the Verde River basin, both feeding the Pardo River. In total, 20 water sampling points were monitored: 17 groundwater monitoring boreholes and 3 reservoirs (Cava da MOU, BNF and Córrego do Consulta) (see Fig. 1). The hydrochemical characteristics of the groundwater were determined using the following techniques:

(a) Piper diagrams: An analysis using Piper diagrams was used to classify the waters and compare the different water groups in terms of the dominant ion.
(b) Species and saturation index: Geochemical simulations were performed using PHREEQE software and were based on the chemical balance concept, which consists of determining the most stable thermodynamic medium through the consideration of all the equilibrium constant systems [2]. The distribution of chemical species or speciation held by PHREEQE takes into account the stoichiometry of formation reactions of each species and the formation of the associated equilibrium constant. The quantities of substances were represented as activities, not as concentrations.
(c) Sulphur isotopes: The samples enriched in 34S present positive values of δ34S, while depleted samples present negative values caused by the isotopic fractioning of sulphur by sulphate reducing bacteria. The isotope 32S forms slightly weaker bonds than 34S. Because of this; the reducing rate of 32S is greater, generating a biogenic sulphide enriched in 32S when compared with the sulphate. Therefore, the low isotopic values are justified by the bacterial reduction of the sulphates [3, 4].

3. RESULTS AND DISCUSSION

The groundwaters classified by the Piper diagram were as follows:

(a) Containing calcium sulphate: P1, P10, P33, P34, P35, P36, P38, P39, P40, P41, P42, P44, Cava, BNF;
(b) Containing mixed sulphates: P16, P30, P36;
(c) Containing sodium bicarbonate: P19, P37, P43, Consulta Creek.

The species of Ca, K, Na, Si, Mg, Mn, Zn, Y, Sr and Cd occur primarily in free ionic form, followed by the sulphated form. Uranium occurs mostly in the oxidized form (U^{6+}) in the species uranyl sulphate and free uranyl. The uranium in the reduced form (U^{4+}) takes the form of hydroxide and fluoride. The relationship between U^{6+} and U^{4+} indicates that the mobility of uranium in the solution is mainly influenced by the redox potential of the solution. The more oxidizing the medium, the higher the ratio of U^{6+} to U^{4+} and thus the higher the mobility in the uranium solution [5, 6].

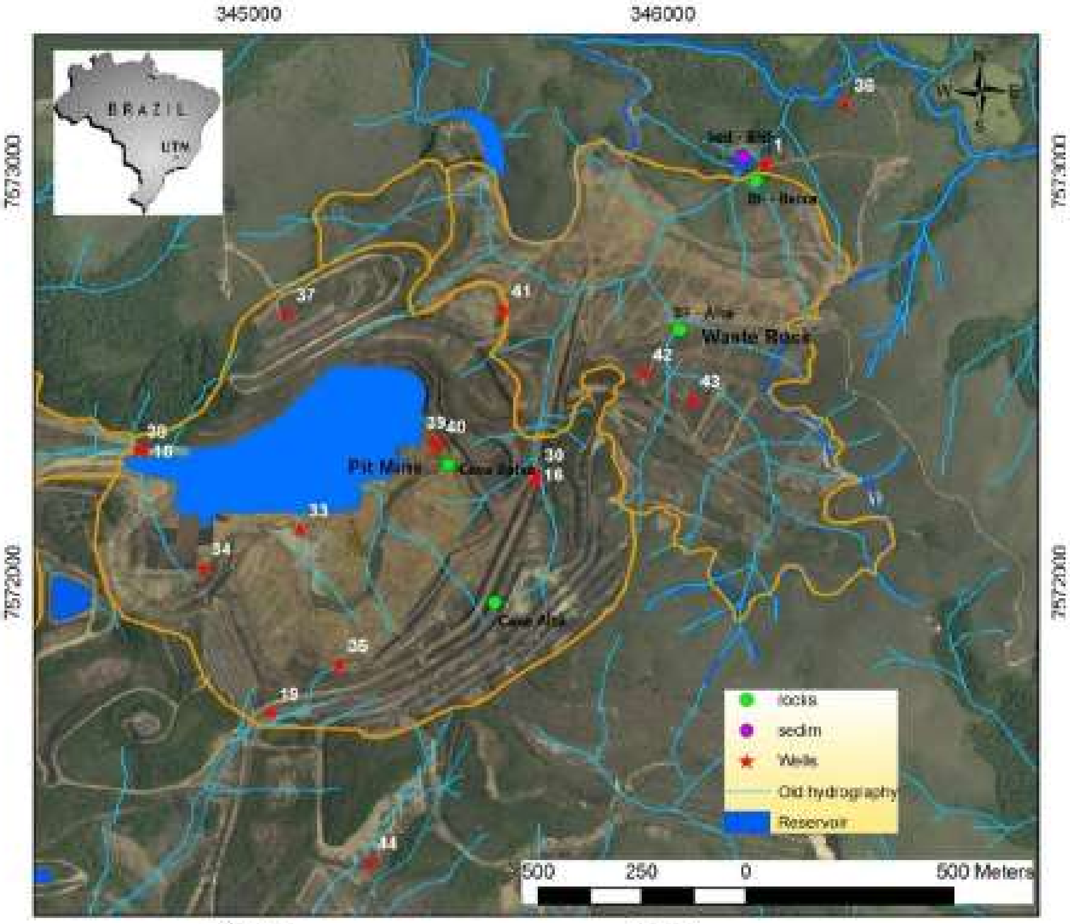

FIG. 1. Location of the study area and sampling water points.

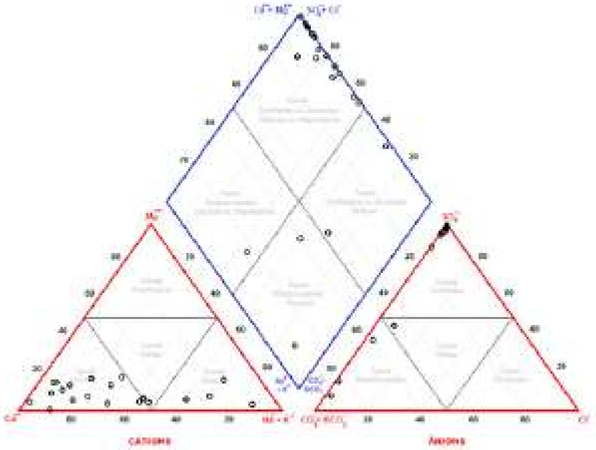

FIG. 2. Piper diagram.

The dominant processes are:

(a) The dissolution of K-feldspar, smectite, ilitas, albite, pitchblende, fluorite and pyrolusite;
(b) The precipitation of muscovite, haematite and kaolinite;
(c) Pyrite oxidation;

(d) Equilibrium solubility of barite, kaolinite, goethite and gibbsite.

The values of $\delta^{34}S - SO_4$ in the monitoring well samples and in the reservoirs vary only slightly and present values between –3.06 and –0.31‰.The results demonstrate that the sulphate present in the waters did not participate in the repeated processes of bacterial oxy–reduction that would result in lower values of $\delta^{34}S - SO_4$. The water and the oxygen show large differences in the isotopic composition of the oxygen. While for atmospheric oxygen $\delta^{18}O$ = 23.5‰, the isotopic composition of the water is typically negative, in this case varying between approximately –12 and –4‰. In this way, it is possible to determine the water and oxygen molecular fraction incorporated in the sulphate [7, 8]. Also, the oxidation of the pyrite due to the molar fraction of the oxygen present in the water, in the average of the wells, was X = 0.7, while the oxidation of the pyrite by oxygen in the air, in the average of the wells, was X = 0.3. The values of $\delta^{18}O - SO_4$ vary between –4.4 and –1.0‰, demonstrating an isotopic signature closest to that of oxygen in the water. The results prove that the oxidation of the pyrite due to the molar fraction of the oxygen present in the water is greater than the oxidation of the pyrite through oxygen in the air.

4. CONCLUSIONS

This study has:

(i) Demonstrated that the groundwater is calcic and sulphated;
(ii) Identified the main chemical elements in solution and their ionic form;
(iii) Identified the dissolved and precipitated major minerals;
(iv) Evaluated the mechanisms of pyrite oxidation (air and/or water).

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Poster Presentation

TREATMENT OF COAL MINING TAILINGS TO REDUCE THE LEACHING OF METALS INCLUDING URANIUM AND THORIUM

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Abstract

Tailings from a coal residue facility in Santa Catarina State, Brazil were treated by separation of the particles according to their density, generating heavy, mixed and light fractions. These three fractions had high, intermediate and low pyrite contents, respectively. The aim of the study was to evaluate the mixed fraction. Leaching from this fraction was evaluated by adding water at a ratio of 4 mL of water to 1 g of tailings. Then, the water–tailings mixtures were centrifuged and filtered for several different time periods. After each period, the leachates were analyzed for pH and electrical conductivity. The concentrations of Al, Fe, Zn, V, Cu, Pb, Cd, Ni, Cr, Mn, As, Ba, U and Th were carried out by ICP methods. The results showed that the pH decreased from 7 (water) to 3.5 (leachate) in the first step, probably because of the leaching of pyrite. Both pH and conductivity increased with contact time and a pH of 5 was attained after 72 h. The metals V, Cu, Pb, Cd, Cr, As, U and Th were not detected in the leachate. Metals showed distinct kinetic leaching profiles. In comparison with other studies, the metal levels and the pH were both below what was observed in impacted local freshwater. Thus, although preliminary, these results indicate the effectiveness of the treatment.

1. INTRODUCTION

Although coal fired power plants account for less than 2% of Brazilian energy production, they generate residues with high pyrite contents, the oxidation of which triggers acid mine drainage, causing the leaching of metals from the residues followed by contamination of the surrounding soil and water bodies. In addition, it is known that certain activities involving minerals (including the coal industry) can give rise to significantly enhanced exposures to radionuclides of natural origin. This may result in the need for monitoring and control for long periods. As part of an investigation to identify a disposal method that does not entail this long term monitoring and control, samples from a coal residue facility situated in Santa Catarina State, Brazil (see Fig. 1) were collected in accordance with NBR 10.007 [1] and treated at Centro de Tecnologia Mineral (CETEM).

2. METHOD

At CETEM, the samples were treated to separate the particles according to their density, generating heavy, mixed and light fractions. These three fractions had high, intermediate and low pyrite contents, respectively. The mixed fraction was not only the largest but is also representative of the one that will be disposed of on land. The leaching of metals was evaluated as shown in Fig. 2. Samples were prepared by adding water to the mixed waste fraction with water type 2, in triplicate, at a ratio of 4 mL of water to 1 g of tailings. The water–residue mixtures were agitated in an orbital shaker (150 rpm) for intervals of 1, 24, 48 and 72 h. At the end of each interval the mixture was centrifuged for 5 min at 5000 rpm and then passed through a 0.45 µm filter. The solutes were discarded while the percolated liquid was analysed immediately for pH and electrical conductivity before being stored in a refrigerator for subsequent chemical analysis. The concentrations of Al, Fe, Zn, V, Cu, Pb, Cd, Ni, Cr, Mn, As and Ba were determined by ICP-OES, while the concentrations of U and Th were determined by ICP-MS.

FIG. 1. Coal residue facility.

FIG. 2. Leaching method diagram.

3. RESULTS AND DISCUSSION

The preliminary results were evaluated against criteria specified in the Brazilian environmental legislation CONAMA Resolution 357 [2]. As expected, the pH and electrical conductivity both increased with contact time (Fig. 3), but the pH levels nevertheless remained low, at 4.95. The leachate could therefore be characterized as an acid effluent, being outside the pH range of 6–9 specified in the legislation for freshwater bodies.

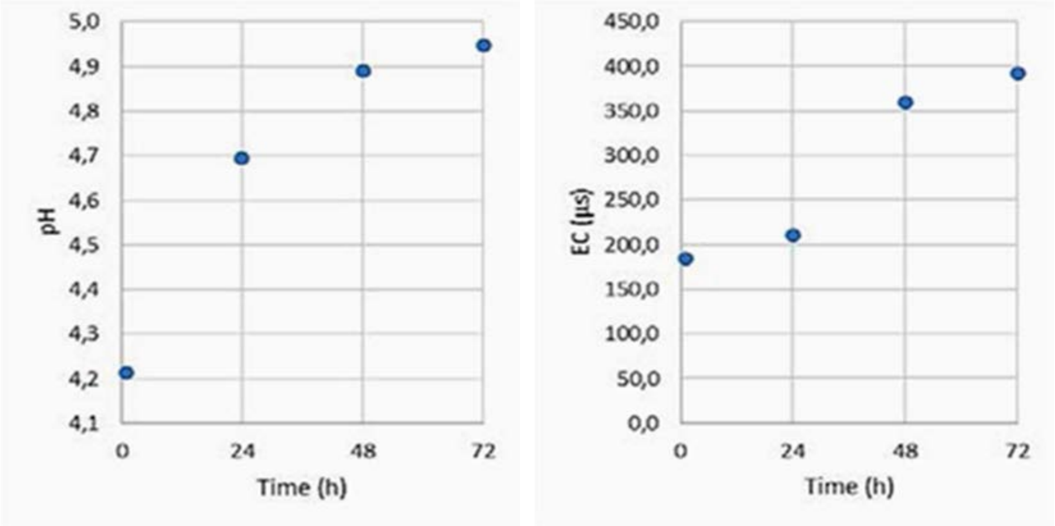

FIG. 3. Results of leaching tests for pH and electrical conductivity.

The chemical results indicated that there are distinct kinetic leaching profiles for each metal/metalloid, as shown in Fig. 4. The elements V, Cu, Pb, Cd, Cr, As, U and Th were not detected in the leachate. All Ba concentrations were within limits. Although Al and Fe concentrations declined with increasing mixture contact time, high concentrations of Fe — exceeding the limits — were still detected after the last interval. The Al concentration exceeded the limit only in the first hour. Concentrations of Ni and Mn presented unacceptable values in every interval in terms of the legislation. The Zn concentration also remained above the limits except after the final interval.

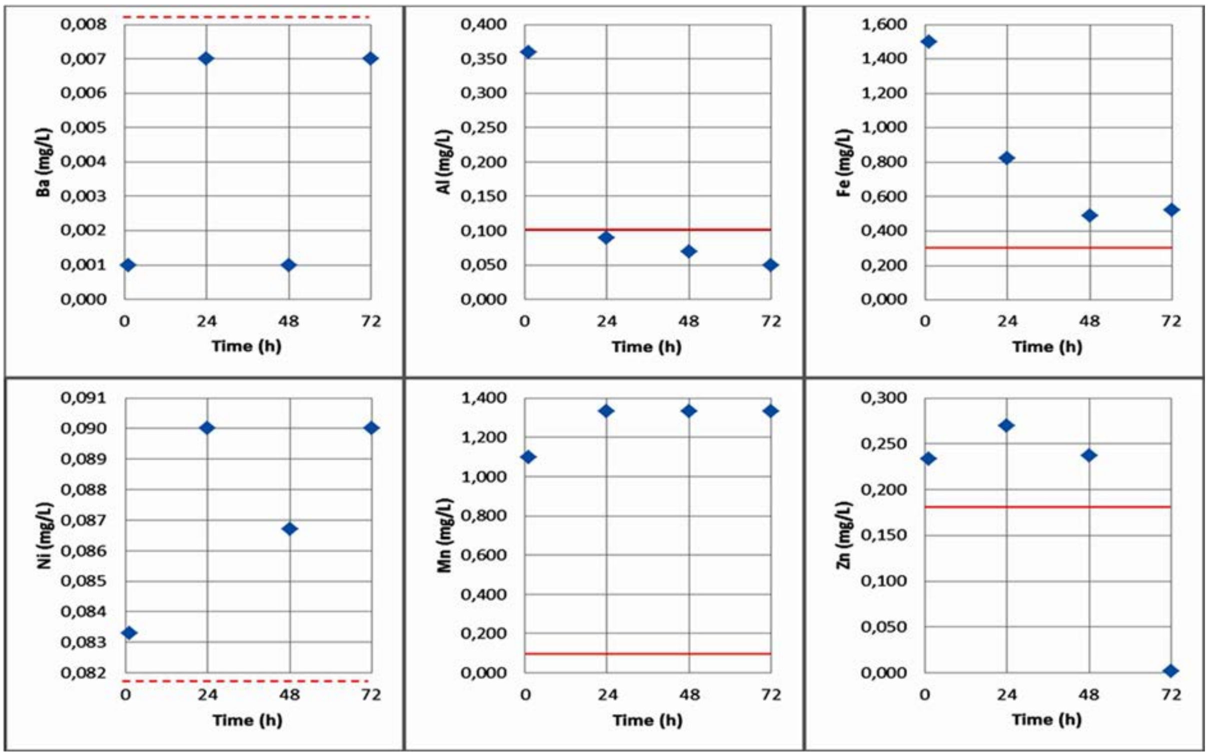

FIG. 4. Results of leaching tests for metals (CONAMA Resolution 357 criteria shown in red).

4. CONCLUSIONS

Although preliminary, these results indicate the effectiveness of the treatment. In comparison with the findings of other studies [3], the metal concentrations and pH are both below the levels observed in impacted local freshwater.

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Poster Presentation

ACUTE TOXICITY OF MANGANESE TO *Ceriodaphnia silvestrii* AND *Daphnia magna* IN BIOASSAYS AND THE POTENTIAL TOXICITY OF THIS METAL IN URANIUM MINE EFFLUENT

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Abstract

Treated effluents from a uranium mine (the Ore Treatment Unit of Brazilian Nuclear Industries) containing acid mine drainage are continuously discharged into the Antas Reservoir and studies have shown that one of the main problems of these effluents released on the catchment basin of the Ribeirão das Antas is their association with high manganese levels recorded in water samples. In this study the effects of acute manganese toxicity to *Daphnia magna* and *Ceriodaphnia silvestrii* were determined. The results were then evaluated against manganese values recorded in the Antas reservoir. The acute 48 h LC50 values for manganese to *C. silvestrii* and *D. magna* were 5.93 and 51.66 mg/L, respectively ($P<0.05$). According to the literature, manganese concentrations recorded in water samples (1.04 to 20.3 mg/L) revealed toxicity potential to *C. silvestrii*. Since the manganese occurs in the composition of the effluent, which may contain other stable and radioactive elements, it is suggested that ecotoxicological monitoring in water samples from the Antas Reservoir should be continued, with the aim of assessing the potential synergistic and antagonistic effects of the chemical mixture that makes up the radioactive effluent that is treated and released into this reservoir.

1. INTRODUCTION

Continuous release of acid mine drainage from mining activities into the environment can cause severe degradation of water quality and other environmental pollution problems, mainly due to the large volume of effluents generated, and can adversely affect aquatic biota. Thus, the formation of this acid mixture, consisting of a large spectrum of chemical pollutants (e.g. Fe, Cd, Cu, Pb, Zn, U, Mn) creates a harsh environment that is harmful to aquatic organisms living around the mining area [1, 2].

Treated effluents containing acid mine drainage from a uranium mine, the Ore Treatment Unit (UTM) of the company Brazilian Nuclear Industries, are continuously discharged into the Antas reservoir in the catchment basin of the Ribeirão das Antas. Studies have shown that that these effluents are associated with the high manganese values of 1.04 to 20.3 mg/L recorded in water samples [3–6] and 16 000 mg/kg recorded in sediment samples [6], often exceeding criteria established in Brazilian legislation (CONAMA 357/2005). An evaluation of the water quality in the Antas reservoir has identified seasonal variations in contamination by fluoride, sulphate, manganese and uranium due to the discharge of effluent from the mine [4]. According to Ref. [7], manganese is a toxic element frequently overlooked in the assessment of effluent toxicity, even though it is a common contaminant in discharges from mining activities.

Brazilian effluent legislation (CONAMA 430/2011) requires that the effluent from a source of pollution must not cause or potentially cause toxic effects in aquatic organisms in the receiving water body.

A study on toxicity assessment of manganese in soft and slightly acidic waters of Magela Creek in Australia concluded that the freshwater species tested presented a broad range of sensitivities to manganese. *Moinadaphnia macleayi* (cladoceran), *Americana cumingi* (aquatic snail) and *Hydra viridissima* (hydra) were especially sensitive, with all IC10 values being lower than 610 µg/L [8]. This finding was probably related to the low concentrations of Ca^{2+} in the natural waters of Magela Creek.

As relatively little attention has been directed to the toxicity of manganese for freshwater organisms, the present study assessed the effects of acute manganese toxicity by means of a 48 h immobilization test, using *Ceriodaphnia silvestrii* and *Daphnia magna* as test organisms. The results of the toxicity tests were then evaluated against manganese values recorded in previous years in the treated effluent from the UTM.

2. MATERIALS AND METHOD

Stock cultures of the cladoceran *Ceriodaphnia silvestrii* [9] and *Daphnia magna* [10, 11] were maintained in continuous parthenogenetic reproduction. Stock cultures of green algae *Raphidocelis subcapitata* were maintained in culture medium CHU-12 [12]. *C. silvestrii* and *D. magna* were fed with suspensions of the alga *R. subcapitata*. For *C. silvestrii*, an algal suspension concentration ranging from 1×10^5 to 5×10^5 cells/mL per organism was provided, while for *D. magna,* the concentration provided at each renewal was 1×10^6 cells/mL per adult organism.

To conduct the tests, manganese solutions were prepared from a stock solution of $MnCl_2{\cdot}4H_2O$ (Sigma-Aldrich®) with a targeted nominal manganese concentration of 1000 mg/L. The dilution water used in the preparation of the test solutions was the same as that used in the maintenance of test organisms (reconstituted water). Preliminary tests were performed using the concentrations of manganese proposed in Ref. [7]. After the preliminary tests the stock solution was then diluted to produce seven manganese concentrations for *C. silvestrii* (3, 4, 5, 6, 7, 8 and 9 mg/L) and *D. magna* (30, 40, 50, 60, 70 and 80 mg/L). The highest and lowest tested manganese concentrations were measured analytically at the beginning of the test and after 48 h, so as to confirm the nominal concentrations tested. For *D. magna* the tests with the divalent metal (as $MnCl_2$) were performed according to ISO6341 [13].

In the tests of acute toxicity over 48 h using different manganese concentrations, neonates of *C. silvestrii* and *D. magna* <24 h old were exposed to manganese concentrations and to control treatment (reconstituted water only). On the acute tests, five individuals in four replicates were used, disposed in polypropylene beakers (Brand®) of 10 mL for *C. silvestrii* and 50 mL for *D. magna*, which contained the solution of interest. The tests were carried out in an incubator with a photoperiod of 16 h of light and 8 h of darkness and controlled temperature (25 ± 2°C for *C. silvestrii* and 20 ± 2°C for *D. magna*), without test-organism feeding throughout the 48 h period. Next, the numbers of mobile and immobile organisms in each tested sample were assessed. Some parameters such as pH, electrical conductivity and dissolved oxygen were measured at the beginning and conclusion of the test. From the test results, the mean LC50 effective concentrations values were determined for both species.

3. RESULTS AND DISCUSSION

According to the acute toxicity test results, the 48 h LC50 manganese values for *C. silvestrii* and *D. magna* were 5.93 and 51.66 mg/L, respectively, a significant difference

($P<0.05$, Tukey test). This may be related to the water hardness dilution value of *D. magna* (hard water, 175–225 mg/L $CaCO_3$), which was greater than the dilution value of *C. silvestrii* (soft water, 40–48 mg/L $CaCO_3$), possibly presenting a more pronounced protective effect on *D. magna*. In Ref. [7], for instance, it was found that the tolerance of *Ceriodaphnia dubia* to manganese was related to water hardness — in that study, no significant difference was observed between moderately hard water (92 mg/L) and hard water (184 mg/L) but the tolerance was significantly lower in soft water (26 mg/L), with acute LC50 values for *C. dubia* averaging 14.5, 15.2 and 6.2 mg/L, respectively. According to Ref. [1], the decrease in toxicity of the metal with increasing water hardness is due to competition between the metal and Ca^{2+} and Mg^{2+} ions for binding sites on cell surfaces of the organisms. When considering the very much lower LC50 value of manganese for *C. silvestrii* (5.93 mg/L) compared with that for *D. magna* (51.66 mg/L), it should be noted that *D. magna* is an exotic species from regions of high hardness freshwaters while *C. silvestrii* is a species native to tropical environments, where water hardness values are commonly classified as soft. This underlines the importance of using species from tropical environments, such as *C. silvestrii,* as reference organisms in toxicity tests, since this species may better represent the toxicity of samples in tropical regions.

According to Ref. [6], which evaluated the chemical quality of the Ribeirão das Antas and Ribeirão de Caldas micro-basins, high concentrations of manganese along with other constituents (fluoride, sulphate, zinc and uranium) were observed in water and sediment samples from the Águas Claras basin (Antas reservoir). Samples from a location that received treated effluent from the UTM presented high manganese concentrations in water (3.6 mg/L) and sediment (16 000 mg/kg), indicating a negative influence of the UTM acid water treatment system over the chemical quality of Ribeirão das Antas. The manganese concentration in the water (3.6 mg/L) was similar to the 48 h LC50 value of manganese observed for *C. silvestrii* (5.93 mg/L), indicating possible toxic effects over the zooplanktonic community of the system. Moreover, the high manganese concentration registered in the sediment (16 000 mg/kg) should be considered since it can affect the aquatic community through availability of the element in the water column or even through the feeding diet of the organisms by ingestion of particles associated with the metal, evidencing the toxic potential of this metal to both zooplankton species in this study. Thus, the authors suggest that additional tests be carried out in sediment samples to confirm the potential toxicity of manganese to freshwater species.

In a study of the daily variations in water quality at Antas reservoir, where the UTM treated effluent was discharged [5], the highest peak values of manganese were at 12:00 (18.2 mg/L) and 18:00 (20.3 mg/L) in the month of February. Given the manganese toxicity potential towards *C. silvestrii* (LC50 = 5.93 mg/L) and since the effluent may contain other elements with seasonal variations, it is suggested that ecotoxicological monitoring in samples of water in the Antas reservoir be performed, aimed at the assessment of potential synergistic and antagonistic effects of the mixture of radioactive and non-radioactive elements in the treated effluent discharged into this reservoir.

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