

Environmental Contamination from Uranium Production Facilities and their Remediation

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ENVIRONMENTAL
CONTAMINATION FROM
URANIUM PRODUCTION
FACILITIES AND THEIR
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FOREWORD

The legacies of past uranium mining and milling activities continue to be a cause of concern and require assessment and remedial action. This problem has been recognized in many parts of the world over the past three decades, but has received special attention since the end of the Cold War.

The larger uranium producers have expended considerable effort and resources in dealing with this legacy. However, it has to be noted that the search for uranium covered almost all countries in the world. The result in some countries is a legacy of numerous small scale mines and mills. For economic and other reasons, including less stringent environmental standards and lack of awareness at the time, these operations may not have been properly closed out and made safe from both a radiological as well as a general safety point of view.

It was thought that countries with this type of uranium mining and milling legacy would benefit from an exchange of views and experiences. The remediation strategies and techniques developed by countries with major problems such as the United States of America or Germany are often not on the scale of the problems in other countries and could not be implemented in those countries. Therefore, this international workshop was organized in Lisbon from 11 to 13 February 2004. This workshop marked the successful completion of an IAEA technical cooperation project designed to assist Portugal in dealing with its uranium mining legacy of over 50 small sites. The workshop provided a forum for sharing the experience gathered during this project.

The IAEA would like to thank F. Carvalho and the Instituto Tecnológico e Nuclear, Sacavém, Portugal, for organizing and hosting the workshop. The Institut de radioprotection et de sûreté nucléaire and COGEMA, France, as well as ENUSA, Spain, are thanked for providing relevant expertise. D. Read (Enterpris, University of Reading) reviewed the manuscript. The IAEA officer responsible for the project and for this publication was W.E. Falck of the Division of Nuclear Fuel Cycle and Waste Technology.

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1. INTRODUCTION

The special risks associated with uranium mining were implicitly evident as long ago as the early 16th century when workers in the silver mines of central Europe, that are now known to contain uranium mineralization, appeared to be more susceptible to pulmonary disorders than workers at other mines. Concerns regarding operational practices are usually greater at uranium mines than elsewhere, although the focus is on situations where workers are in immediate contact with the ore and processing streams. Over the past decades, these concerns for worker health and safety have been the driving force for gradually tighter controls and improved practices. Only in relatively recent times, however, have concerns been voiced over impacts on public health and on the wider natural environment from the full range of operational activities related to uranium mining and the rest of the nuclear fuel cycle. In the case of the natural environment, these concerns include the risk of environmental degradation, contamination, reduced ecosystem viability and biodiversity, aesthetics, public amenities, access to land, and quarantining of land for future beneficial land use.

Uranium mill tailings are of particular environmental concern because:

- (a) They retain much of the radioactivity of the ore from which they were derived;
- (b) Their radioactivity is very long lived;
- (c) They contain a range of biotoxic heavy metals and other compounds;
- (d) They may contain sulphide minerals which can generate acid mine drainage;
- (e) Their granular to slime consistency makes them susceptible to leaching, erosion or collapse under various conditions;
- (f) The common method of surface disposal exposes a large surface area to the natural elements and thus increases the risk of release of radiation flux, radioactive and geochemically toxic dusts, and interaction with surface water systems;
- (g) The large surface area of these generally thin tailings deposits (or 'piles') adversely affects large areas of land and renders potentially valuable land unfit for other uses.

2. HISTORICAL DEVELOPMENTS

Around the middle of the 20th century a concerted effort was made to discover and exploit uranium resources to supply uranium for the production

of military weapons. Development commenced during World War Two and continued through to the 1960s. This urgency fuelled the first ‘uranium rush’; for example, the governments of the United States of America and the United Kingdom stimulated exploration for uranium by cash rewards for new discoveries. In other countries, exploration as well as development was largely in government hands in order to match the buildup of uranium materials and weapons stockpiles for strategic military purposes. Notable in this period were the:

- (a) Rapid development of many new mining districts (or enlargement of some areas previously worked for small quantities of uranium and radium);
- (b) Development of regulatory frameworks focused on worker health;
- (c) Tailings disposal practices that generally saw tailings placed adjacent to mills, using practices common to other metal mill tailings of the day — i.e. no design features to improve containment security, reduce radon flux, isolate the tailings from wind or water erosion or interaction with surface and groundwater systems;
- (d) The small size of many mines and the level of government control saw many of the mines serviced by central mills, thus focusing tailings disposal problems in relatively few areas that (for reasons of transport, employment, accommodation and servicing) were often close to urban centres.

The first uranium rush had subsided by the end of the 1960s but the potential of nuclear power for peaceful energy generation had been recognized. Concerns over the limits of oil supplies for the world’s energy needs led to projections for substantial increases in the then depressed price of uranium, so that from the mid-1960s to the mid-1970s a second ‘uranium rush’ occurred. This differed significantly from the previous rush in that:

- (1) The impetus was driven by market forces and the world commodity market;
- (2) A range of much larger uranium deposits was discovered;
- (3) Economies of scale at these larger mines and their extended profitable lifetimes allowed more thorough planning of facilities, including tailings facilities, and integration of mines with dedicated mills;
- (4) Lower production costs in association with the downturn in uranium prices at the end of this period forced the closure of many of the smaller mines developed in the first rush.

The last ‘age’ of uranium mining coincides with the period of environmental awareness which began in the mid-1970s. This awareness was born out of a recognition of the level and types of impacts that human activity, including mining, was having on the environment. In particular, this period coincided with:

- (i) Development of the first set of national environmental regulations in several key uranium producing countries;
- (ii) Introduction of legislation for and conduct of the first environmental impact assessments for major mining operations;
- (iii) Public health risks from the nuclear fuel cycle becoming more widely known;
- (iv) Concern for possible impacts upon the natural environment;
- (v) Environmental impacts from the previous generation of uranium mines becoming better understood.

This still continuing age has also been a period of major research efforts into the environmental issues related to uranium mining, and to uranium mill tailings in particular. Research has continued at a high level since the late 1970s. Advances have generally been uneven between different countries because of the economic value of the uranium resource; the level of public and regulatory concern; and the levels of funding made available for technology development.

It must be said that the ages described above have not occurred over the same periods in different countries. The varying levels of strategic importance given to uranium as an energy and weapons feedstock; varying times of opening up to and participation in a free market for uranium; and varying levels of secrecy and centralized control of the uranium mining and nuclear sectors, has meant that many countries have progressed through these ages at considerably slower rates.

3. MINING AND MILLING FACILITIES

In general terms older uranium mining involved ore extraction from a large number of small workings, usually shallow excavations clustered in ‘uranium districts’. It was common practice to have only one mill processing the ore from each district, and in some situations a mill would also treat ore from outlying mining districts as they were discovered. This trend was reinforced by individual mining companies often being too small to afford to build a processing plant of their own; of mines not having sufficiently long lives to warrant the cost of constructing dedicated processing plants; and tight

government control on the production of uranium oxide because of its high strategic importance. It was not unusual for these 'central' mills to be built close to towns in order to benefit from transport and industrial infrastructure, easy access of workers and suitable accommodation for them.

The practice at almost every uranium processing plant is to dispose of the tailings at the nearest convenient place. Therefore the approach of processing uranium ore at central mills close to towns had the unfortunate consequence of placement of tailings close to populated areas where the risk of disturbance and exposure to the general population was highest.

4. SOURCES OF HUMAN HEALTH RISK AND ENVIRONMENTAL IMPACT

4.1. RISKS FROM MINES

There are two basic types of mine, open cast and underground. Each type has specific risks associated with it, but some are common to both. Any excavation, whether underground or open cast, is subject to degradation and may eventually collapse or cave in. Open cast workings are subject to normal erosion forces leading to, for example, slope instability and failure of rock walls, with the ensuing risks of injury and damage to adjacent property. The collapse of underground workings may cause damage at the surface to property and possible injury to the local population. Unsealed mine shafts pose a risk of accidents to unsuspecting forest workers, hikers, etc. Abandoned adits might invite people to enter and explore them, being unaware of the unsafe conditions.

Mine workings frequently extend below the groundwater table, thus providing direct pathways for interaction with the atmosphere. The introduction of oxygen and resulting oxidation of sulphide minerals in the ore or host rock will lead to the generation of acid mine drainage. This is a common phenomenon and is not specific to uranium mining. The effects of acid drainage are discussed in the following section. In turn, direct access to groundwaters also opens up pathways for the introduction of contaminants into the groundwater.

The mining operations themselves may have introduced contaminants into the (aqueous) environment. Examples include degradation products of explosives, machinery oil or transformer fluids.

4.2. RISKS FROM RESIDUE MANAGEMENT FACILITIES

The causes of acute failure in uranium mining and milling residue impoundments are no different from those at other mines. The causes include: earthquake induced instability and failure owing to cracking or liquefaction; physical weakness of embankments and slopes leading to breaching; erosion from heavy rain or adjacent waterways leading to dispersal of residues and thinning of retaining dams or overtopping by decant water; cracking induced by settlement; piping; slumping of material into the containment causing overtopping and/or erosion of the wall or its physical destruction; and spillway collapse caused by heavy overflow of decant or slurry after severe rainfall.

There are also many forms of chronic failure. These include:

- (a) Dispersal of radioactive dust — uncovered residues are allowed to dry out, followed by wind blown distribution of dust;
- (b) Erosion of residues from the outer surfaces of the containment;
- (c) Acid drainage through the floor and/or walls of the containment;
- (d) Contaminated surface runoff draining into natural waterways.

Both acute and chronic events are commonly associated with non-radiological impacts related to heavy metals and other toxic compounds in the tailings, pore water and decant water. The generation of acid drainage from residues containing sulphide minerals is of particular concern. Acid drainage tends to exacerbate the probability of environmental contamination because of the solubility (and hence greater mobility) of many metals under acidic conditions. It may also affect the bioavailability of toxic metals and compounds. Hence the neutralization of tailings prior to deposition, when required, has become common practice in more recent times. Surface capping of solid residues to prevent the ingress of seepage waters helps to reduce the generation of acid drainage.

4.3. HUMAN HEALTH RISKS

Sudden failure of any engineered structure may cause death through drowning, crushing or suffocation. Uranium mining and milling residue containments are no different from water containment dams in that regard. Landslides involving uranium mining and milling residues have killed people and destroyed property. However, the failure of uranium tailings containments does not feature highly on the list of dam failures to have caused death or major

destruction, perhaps because dams at uranium mines represent a small proportion of the total number of mine dams.

Although other types of mining residues may also contain naturally occurring radioactive materials the particular human health risk that is associated with uranium mill tailings is the risk from radioactivity. In turn uranium mill tailings may also pose risks associated with their inventory of heavy metals and other chemical elements, such as arsenic. People in uranium mining districts may be exposed to radiation doses from mining, milling, transport of radioactive materials, radioactive dust and contaminated water and foodstuffs.

Tailings may be a significant proportion of the health hazard because of the manner in which they were commonly disposed of. Their disposal at the surface over relatively large areas allows significant flux of radioactive gases and interaction with surface water. In many historical uranium mining areas both have constituted a pathway from the immediate surroundings of the mine site and affected the general public.

The long half-lives of radionuclides from uranium tailings and the demonstrated risks associated with them have given rise to high levels of concern among the general public and in government — in some places exacerbated by official secrecy and lack of data on health impacts.

Perhaps the most direct implication of mining and milling residues as a radiation source sufficient to cause human health impacts relates to their re-use for building materials. This has happened in many places around the world.

4.4. ENVIRONMENTAL IMPACTS

Many articles in the literature purport to describe impacts on the natural environment from uranium mining in general, and the placement/disposal and management/mismanagement of mill tailings in particular. However, the description of environmental impact that most of these articles present is limited to a qualitative description of impact in terms of contaminated soil, surface waters, groundwaters, lake sediments, etc. Any quantitative description is usually given in terms of the area/volumes affected and concentrations of radioactive elements, or activity levels and doses. A body of literature exists on the contamination of environmental media directly linked to consumption and therefore potential uptake of radionuclides by humans, principally soils and water. In contrast there are relatively few articles that set out to quantify the level of harm to the biosphere caused by uranium mining and milling residues.

Research into the biological impacts of the nuclear fuel cycle is a relatively new science and is strongly biased towards research on the risks,

hazards and impacts to humans from the fuel fabrication, power generation and fuel reprocessing/disposal parts of the cycle. However, interest in research into the biological impacts from the uranium production part of the nuclear fuel cycle has grown from:

- (a) A realization that fisheries in the vicinity of some uranium mines were deteriorating;
- (b) A realization that radionuclides could be taken up and concentrated by plants, thereby posing a potential risk to vegetation as well as to animals higher in the food chain, including humans;
- (c) A realization that radionuclides could be taken up and concentrated by animals, thereby posing a potential risk to the health of individuals, communities and ecosystems as well as animals higher in the food chain, including humans;
- (d) Documentation on the concentration of radionuclides in bottom sediments and their long term availability to bottom feeders and potential for multiple cycling through the aquatic food chain over the long term;
- (e) Development of transfer pathway models for the transfer of radiological dose through the food chain;
- (f) Concern that tangible impacts on human health from radioactive contamination associated with uranium mines and tailings piles may also be having significant impacts on the environment;
- (g) Questions over the validity of the ICRP's assumption that if human beings were adequately protected other species would also be protected.

Since the late 1970s there has been a general increase in concern for the environment, presumably due to evidence that the actions of humans are causing visible and significant environmental changes. Furthermore, close attention is now being paid to the larger implications of harm being caused to elements of particular ecosystems with consequent impacts on biodiversity. It is perhaps for these reasons that the issue of environmental protection in the context of ionizing radiation is being addressed in many countries.

The impacts on the environment from uranium mining and milling residues are not all related to the radionuclide content alone. The presence of other contaminants can exacerbate the availability of the radionuclides to the environment, and in some cases the other contaminants have direct harmful effects in themselves. In this regard, the potential for uranium mining and milling residues to cause environmental harm is little different from that of other forms of mining, and the resultant impacts may be quite similar. Indeed it is not adequate to consider the radiological risk only. The other effects may include:

- (1) The chemical toxicity of the radionuclides, including uranium;
- (2) The chemical toxicity of heavy metals and metallic compounds;
- (3) The chemical toxicity of non-metallic minerals and compounds in the ore or introduced during processing (e.g. sulphuric acid, kerosene);
- (4) Acidity, resulting from sulphidic (ore) minerals or acid introduced during milling;
- (5) Increased turbidity in surface waters;
- (6) Increased salinity.

The types of non-radiological contaminants that may cause harm are dependent on the mineralization in the ore body, the gangue mineralogy, the overburden mineralogy and the processing technique used in the mill. Elevated acidity plays a major role in increasing the mobility of metals in aqueous solution, including uranium, as well as copper, arsenic, cadmium and other metals. The effects of heavy metal pollution are complex and strongly dependent on local geographic and climatic factors, the mix of chemical constituents present, and on the nature of the affected organisms. Effects can be lethal or non-lethal. Chronic sub-lethal poisoning can affect growth, reproduction, behavioural patterns, and result in lower resistance to disease, as well as causing the organism to have body concentrations of elements above standards permitted for consumption. In some instances the non-radiological hazards have a much larger effect than the radiological contaminants.

5. ADDRESSING THE URANIUM MINING LEGACY

The incentive for remedial work to be undertaken is compliance with regulatory requirements. Examples of remediation of historic uranium sites by the private sector are limited. Many of the mines concerned were operated by government agencies or were subject to tight government control through to their closure. In the last twenty years several governments around the world have implemented major remedial work. The timing and type of programme have been influenced by several different factors. These include access to information on health and environmental effects, changes in the attitudes of government agencies towards making information available to the public, access to sufficient funds, the type and extent of operations, and their impacts on the environment. Funding is not necessarily an impediment in Western countries, provided that the problems translate into political debate and pressure. However, some other countries have been unable to implement remediation owing to a lack of funding or political willingness. The breakdown of the Soviet Union made public discussion on impacts from uranium mining

and milling possible for the first time in many countries. German unification led to sufficient funding for dealing with the impacts from uranium mining and milling in the former German Democratic Republic. Plans for expansion of the European Union to include Central and Eastern European countries led to the development of a funding scheme for remediation planning and execution in those countries. Similar assistance is being given by the EU to former Soviet Republics.

Addressing the mining legacy typically proceeds according to the following basic steps:

- (a) Assessment of the problem vis-à-vis the regulatory regime;
- (b) Development of a remediation strategy;
- (c) Implementation of remedial actions;
- (d) Compliance monitoring;
- (e) Stewardship, if needed.

6. OBJECTIVES OF THE WORKSHOP

Many countries with smaller scale uranium mining and milling activities have closed down their respective activities. Presently they are in the process of assessing this legacy and may undertake measures to safely close out mines and residue management facilities. In addition, possible environmental impacts are being assessed and remediation measures are being undertaken.

Countries with smaller scale operations, e.g. France, Portugal or Spain, are often in similar geographical, geological and climatological settings. Deposits are of small to medium size, vein type in granite or metamorphic rock and may be widely distributed geographically. Accordingly, mining and milling methods may be similar.

The level of progress in adequately dealing with this legacy is quite varied from country to country, depending on national priorities and resources available. Some countries, such as France, have already subjected all known sites to risk assessment and remedial actions if necessary. Other countries are still trying to understand the size of their respective legacies.

Conferences and symposia on the uranium mining legacy and pertinent mitigation strategies and measures have taken place at regular intervals since the early 1990s. However, they were naturally dominated by those countries with large scale legacies, such as the Czech Republic, Germany and the USA, whose respective strategic and management approaches tend to be proportionately larger. Considering this situation, a forum for exchange for countries with smaller scale uranium mining operations was deemed to be of likely benefit.

7. SUMMARY AND CONCLUSIONS

Uranium exploration, as well as mining and milling, have taken place in many places around Europe. The small exploitable reserves often did not lead to sustained mining activity, but nevertheless may have resulted in radiological and non-radiological legacies that need to be addressed. Today's regulatory and licensing procedures, that take the entire life cycle of a facility into account, aim at preventing such legacies from occurring in the first place and the creation of new legacies as a result of site closure programmes (see paper 6 in the Appendix).

Some of the larger scale mining operations (see paper 20 in the Appendix) have resulted in environmental impacts, but even the geochemical signature of relatively small mining operations can be detected quite a distance away, as is demonstrated in respective field investigations (papers 3 and 14 in the Appendix).

The impacts and risks may be due to the release of contaminants, but a variety of geotechnical risks may also be present. A typical situation is the inadequate siting and geotechnical planning of residue management facilities, as shown in paper 10 of the Appendix.

Further decisions on how to address the legacies need to be based on an adequate understanding of the type and extent of the contamination and of the mechanisms and pathways by which environmental impacts and human exposure might occur (see papers 3 and 14 in the Appendix). This is developed by undertaking an adequate programme of site investigation (see papers 10, 11 and 17 in the Appendix). Deconvoluting the anthropogenic contamination signals from natural background is often not trivial in the case of former uranium mining areas, as shown in paper 15 in the Appendix.

Given the widespread and dispersed occurrence of uranium mining legacies in some countries, a mechanism for identifying the driving forces behind environmental impacts and prioritization of remedial actions may be needed, also taking into account resources becoming increasingly scarce. To aid such processes in a systematic way, various European and other international organizations have developed the so-called DPSIR (driver–pressure–state–impacts–response) framework. The main objective is to help address the driving forces behind rather than symptoms of environmental impacts and to have a (semi-)quantitative tool for assessing cause–effect relationships in a logical framework. paper 1 in the Appendix provides an example of a risk assessment based on the DPSIR framework. While this is an approach that has not been used in such a context yet, care needs to be exercised in selecting the variables. For example the use of 'volume of mining residues' as a 'pressure' indicator may be too simplistic and misleading, as the main pressure does not

necessarily arise from the volumes as such, but from the quality of (long term) management.

In any case the processes are likely to be very complex, resulting in the possible need for extensive site investigation and characterization programmes. In order to focus available resources and optimize expenditure, the relevance of certain parameters with respect to the processes being investigated needs to be assessed. In paper 15 of the Appendix, Pereira et al. present a possible approach, using Monte Carlo simulations as a basis for parameter sensitivity studies. It should be noted, however, that the underlying conceptual model needs to be verified periodically on the basis of available site characterization data.

Managing the uranium mining legacies requires a careful evaluation of the various strategic and technical options. The decision making process can be aided and made more transparent by using quantitative tools. In paper 4 of the Appendix, Deissmann and Kistinger provide an example of the application of such a method, though the monetarization approach is not universally accepted. The main issue here is the appropriate selection of a monetary value for certain variables, which is open to ethical discussion.

The organizational difficulties in implementing even relatively small scale remediation projects have been demonstrated by Ruiz Santos and Sánchez Delgado (paper 18 in the Appendix). The authors also emphasize that preference should be given to 'passive' over 'active' solutions, an observation that is even more relevant given the background need for long term management.

We are now looking back at 15 years of increased remediation of uranium mining and milling legacies. In certain countries, based on the current scientific and technical insights, a first campaign of assessment and remediation where needed has been completed, for example in France. Hence, first 'lessons learnt' and other evaluations of experience gained have become available (papers 8, 13 and 15 in the Appendix). The fact that the regulatory framework and (radiological) remediation criteria had to be developed while actual remediation programmes were already under way has to be acknowledged when judging the decisions and the results.

In summary, it appears that we have sufficient scientific and technical knowledge to assess the environmental impact of uranium mining legacies. For a given case, however, resource constraints may impose limitations on developing a comprehensive understanding of a site. Hence a considerable amount of uncertainty in the validity of a remediation solution may remain, although we would in principle have the capability for adequate scientific and technical solutions. The efficacy of remediation measures to reduce

radiological exposures has been demonstrated in field investigations (see papers 5 and 18 in the Appendix).

A reasonable balance between the scope of the remedial actions and long term stewardship needs, obligations and associated costs has to be found (see paper 16 in the Appendix). As has been pointed out by Falck (paper 7 in the Appendix), the technological and organizational challenges posed by the long term stewardship programmes are reasonably well understood, but there are no satisfactory and 'proven' solutions to many of these problems. The present generation may have to be content to provide state of the art technical solutions while accepting that future generations may have different judgements. Our engineered solutions may well become the contaminated sites of the future.

Appendix

PAPERS PRESENTED AT THE INTERNATIONAL WORKSHOP

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Paper 1

PRELIMINARY RESULTS OF A RISK ASSESSMENT STUDY FOR URANIUM CONTAMINATION IN CENTRAL PORTUGAL

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Abstract

This study applied DPSIR methodology for developing response indicators and establishing the relationship between central and local governments as far as risk management is concerned, and was also based on the findings from an EC project, the European Spatial Planning Observation Network (ESPON) 2006 PROGRAMME Action 1.3.1. In order to apply the appropriate response indicators it is necessary to identify driving forces (D), pressure (P), state (S), and impact (I) indicators. The purpose of this study is to show the application of two methodologies in dealing with the ‘uranium mine contamination’ hazard in central Portugal with the available indicators. The central region of Portugal (NUTS II, Nomenclature des Unites Territoriales

Statistiques, Level II) occupies an area of 23 666 km² (25.7% of Portugal) and comprises 78 municipalities. Its population is almost 1.8 million inhabitants (17.2% of the country's total). The 10 NUTS III areas of central Portugal contain a large number of mineral occurrences, as well as abandoned uranium mines exploited since 1907 and located mainly in granitic rocks. Two different methodologies were applied consecutively: (1) a methodological approach where specific regional indicators were studied per municipality; (2) the ESPON-HAZARDS project methodology, using the identified ESPON-HAZARDS indicators studied per NUTS III region. It was concluded that the risk map created by the application of the regional methodology needed revision. Considering the available indicators in the present scenario the highest risk municipality was Nelas and the possible response indicators for the higher risk areas could be: 'monitoring of water supply' and 'conditioned water and agriculture use'. The ESPON-HAZARDS methodology is applied using the available data, and the achievements indicate that the map of central Portugal needs revision. The reason is that although the presence of the mines per NUTS III is raised, the risk is determined by the type of storage and volume of waste that influences the hazard occurrence according to ESPON definitions [II-2 and II-6]. With the present map the higher risk NUTS III were Dão-Lafões and Pinhal Interior Norte.

1. INTRODUCTION

One of the main tasks of ESPON [1] Project 1.3.1, entitled The Spatial Effects and Management of Natural and Technological Hazards in General and in Relation to Climate Change, is to study and assess the existing and proposed indicators related to natural and technological hazards. An indicator is a tool to measure and indicate the development of various phenomena. It shows the condition of a system and the changes over time [2].

The European Environment Agency (EEA) has proposed indicators to express the condition of complex systems, summarizing the complex message into a manageable and understandable one. Each indicator by itself tells a story as part of the total, and only by combining indicators is it possible to gain a complex view [3]. The ESPON-HAZARDS project and this paper use the EEA's definition for indicators, the DPSIR (driving force, D; pressure, P; state, S; impact, I; response, R) framework [4]. Referring to the risk concept 'driving forces' refer, for example to geophysical processes and climate change, 'pressure' refers to the cause of a risk, 'state' refers to levels of contamination, 'impact' refers to the effects on humans and the environment, and 'response' refers to mitigating actions taken.

The central region of Portugal extends over a 23 666 km² area (25.7% of mainland Portugal) and includes 78 counties. Its population is close to 1.8 million inhabitants (17.2% of Portugal's total), of which 65% is of working

age. The region contains ten NUTS (Nomenclature des Unités Territoriales Statistiques) level III areas [5]. It is traversed by Portugal's main mountain chain, the highest peak being the 'Serra da Estrela' (1991 m). It includes the catchment areas of some of the most important Portuguese and Iberian rivers (Mondego, Vouga, Dão, Lis, and Côa, which are tributaries of the Douro and Zêzere, which is a tributary of the Tejo), and several water resources, including thermal and mineral waters, of the greatest strategic importance for the region and for the country [6].

The 'Orla Ocidental' occupies the central coastal region of Portugal and consists of a sedimentary belt comprising Triassic to Quaternary geological formations. The Central Iberian Zone, southwest sector and part of the Douro-Beiras sector correspond to older Proterozoic formations up to the Carboniferous. Several varieties of granitic formations occur in this Central Iberian Zone, such as Late Hercynian monzonitic granites, that host most of the uranium mineralizations [7].

Uranium exploitation began in central Portugal in 1907 with the Urgeirica ore discovery and continued until 2001 with the closure of the Sevilha and Quinta do Bispo mines. A total of 60 mines were exploited — 45 open pits and 15 underground operations — and 456 mineral occurrences were identified and evaluated during the exploration work. The fact that the waste deposits resulting from uranium mining are different and cause different effects on the environment justifies the selection of a 'uranium mining contamination' hazard [8].

This study is part of the work undertaken under the ESPON HAZARDS project and the objective is to select regional indicators for 'uranium mining contamination in central Portugal' hazards and to compare them with the ESPON indicators to be used at the EU level.

2. METHODOLOGY

2.1. Overview

The first methodological approach, the regional methodology, is represented by the regional DPSIR indicators and, according to their importance, weighting factors were given to these indicators. The vulnerability factors were considered to be the impact indicators due to their relation to the impact on humans. The municipality was used as the classification unit. In the second methodological approach, i.e. the ESPON methodology, the DPSIR indicators were adapted from the regional indicators because the required information to develop the appropriate indicators (for all European countries

in the respective hazard) was not available at the time. The concept of ‘degree of vulnerability’ in the ESPON methodology was introduced, based with equal weight on population density and gross domestic product (GDP) per capita. The classification unit in the ESPON methodology was NUTS III.

2.2. The first methodology approach

The following indicators were selected as driving forces: (1) topographical ‘slope’ (%), and (2) ‘flow accumulation’, meaning an integer number that represents the number of upstream digital elevation model (DEM) cells (calculated with geographic information systems (GIS) from altimetry) whose flow paths ‘pass through’ the given DEM cell. These two indicators were GIS derived layers from altimetry data [9]. The ‘land use’ (3) according to the Inventário Florestal Nacional [10] and ‘lithology’ (4) (Atlas do Ambiente Digital) are also considered as indicators. As pressure indicator, the ‘presence of uranium mines’ (5) was selected from the SIORMINP database [11]. The uranium concentration in stream sediments in the central region, expressed as a ‘geoaccumulation index’ (6) [12] was selected as state indicator [13]. As impact indicators the ‘water systems per municipality’ (7) [14] and ‘inhabitants per water system’ (8) [II-14] were chosen because the impact represents the effect in human lives, although contamination information was not available [14]. Response indicators are the final result of this study.

The justification for classification of the above eight indicators used is explained in the following:

- (1) Slopes of up to 11% were considered low influence factors, 11% to 19% as medium influence factors, and from 19% to 34% as high influence factors. Slopes were obtained from the digital topographical map (DTM) [15] and calculated from the contour elevation shape file.
- (2) The flow accumulation was considered relevant because water is the most important means of transport of uranium contamination.
- (3) Agriculture and surface water reservoirs are considered to be high sensitivity areas for the land use indicator (high risk factor) for contamination by uranium mining. Urban areas were considered of medium sensitivity, while forest, unproductive, and uncultivated areas were considered to be lower sensitivity areas.
- (4) The relevance of the lithology indicator stems from the knowledge that the main occurrence of highly concentrated uranium minerals is related to granitic rocks and to shales in close contact with the granitic intrusions. Three classes were obtained: low risk class: limestones, sandstones, silts,

clays; medium risk class: shales and quartzites; and high risk class: granitic rocks.

- (5) The uranium mining indicator corresponds to the number of uranium mines and occurrences per municipality. The distance from the uranium mines and occurrences to the different land use categories was used as a basis. Uranium mines that are 500 metres away from agricultural land and groundwaters were considered to be of 'high risk', and of low risk where the distance to the forest, unproductive and uncultivated land is 50 km.
- (6) The geoaccumulation index is defined as [12]:

$$I_{geo} = \log 2 [C_n / 1.5 \cdot B_n]$$

where: C_n is the concentration of the chemical element n in fine grained fraction of the sediments concerned; B_n is the geochemical background concentration in the clay fraction of sediments (average clay value). The adjustment factor '1.5' was calculated by the authors of this paper to provide for local lithological effects in the background values.

The results were grouped into seven classes, as shown in Table 1.

- (7) The 'inhabitants per water system' indicator was considered relevant because more systems per municipality imply higher vulnerability. Small systems at small villages are more vulnerable to this kind of contamination because they are often uncontrolled. More systems per municipality also mean smaller systems in each municipality.

TABLE 1. CLASSIFICATION OF THE GEOACCUMULATION INDEX

I_{geo} class	Geoaccumulation index	Degree of contamination
6	>5	Very strong contamination
5	>4-5	Strong to very strong contamination
4	>3-4	Strong contamination
3	>2-3	Moderate to strong contamination
2	>1-2	Moderate contamination
1	>0-1	Weak to moderate contamination
0	<0	No to weak contamination

- (8) The ‘inhabitants per water system’ indicator was considered important due to the fact that water for human consumption offers the highest risk for radioactive contamination. This information was obtained only for groups of supply systems in each municipality. In the future it needs to be developed with more detailed information.

Classification factors to compose the risk map are considered in three classes: (1) low risk factor (dark grey), (2) medium risk factor (lighter grey), and (3) high risk factor (medium dark grey) and are shown in Table 2.

2.3. The ESPON methodology

For application of this methodology to the case of Portugal the hazard ‘hazards from hazardous waste deposits’ was not available and the location of uranium mines and mineral occurrences was used instead. The indicators used were: (1) the pressure indicator ‘number of uranium mines per NUTS III’; and (2) the impact indicator ‘number of water supply systems per NUTS III’, although no contamination information was available. Each indicator was calculated for each NUTS III and classified into one of five classes, I to V. The sum of both classifications was also classified from I to V and in this way the intensity of hazard was obtained (Table 3).

The ‘degree of vulnerability’ reflecting the vulnerability of a region is analysed by (1) the GDP per capita and (2) the population density. These indicators already include a variety of factors that have more or less influence on vulnerability. The GDP per person in combination with the population density stands for the economic damage potential (infrastructure, buildings, movable facilities), the technical response potential and the probable injury to people at the same time. The combination of ‘intensity of hazard’ and ‘degree of vulnerability’ is synthesized in Table 3, which summarizes the hazard vulnerability [2].

3. DISCUSSION

3.1. Factor maps calculated by the first methodology

Factor maps calculated with three classes and the respective multiplication factors are presented in Table 2. In the first case the classification was first made according to three classes of land use categories, followed by the operation (Land use categories \times 10 000)/Distance to uranium mines. The

TABLE 2. FACTOR MAPS FOR THE HAZARD ‘URANIUM MINING CONTAMINATION’















Hazard map		Class	PF
(a) Distance from uranium mines to land use categories 	(b) Geoaccumulation index 	(a) [km]	Multiplication factor of 3
		 0–20	
		20–40	
		40–60	
		(b) [non-dimensional]	
		 0–1	
(c) People per water system per region 	(d) Water system per region 	(c) [number]	Multiplication factor of 2
		 104–5145	
		5145–30351	
		30351–70681	
		(d) [number]	
		 0–1	
(e) Lithology 	(f) Flow accumulation 	(e) rock type	Multiplication factor of 1
		 limestones	
		sandstones, shales, quartzites	
		granites	
		(f) [number]	
		 0–884	
(g) Slope 		(g) [%]	Multiplication factor of 1
		 0–11	
		11–19	
		19–34	

TABLE 3. INTENSITY OF HAZARD OBTAINED FROM REGIONAL INDICATORS FOR THE ‘URANIUM MINING CONTAMINATION’ HAZARD

NUTS III	Water systems/ NUTS III	Class	U mines/ NUTS III	Class	Sum	Intensity of hazard
Beira Interior Norte	176	IV	209	V	9	V
Pinhal Litoral	30	II	0	I	3	II
Pinhal Interior Sul	185	IV	0	I	5	III
Beira Interior Sul	57	II	2	I	3	II
Cova Da Beira	63	III	24	IV	7	IV
Serra Da Estrela	57	II	36	IV	6	III
Dão Lafões	190	IV	76	V	9	V
Pinhal Interior Norte	225	V	27	IV	9	V
Baixo Mondego	36	II	2	I	3	II
Baixo Vouga	59	II	1	I	3	II

resulting factor map was reclassified for three classes and assigned a weighting factor of 3, meaning that this map would count 3 times on the final map.

The second factor map considered the geoaccumulation index, uranium concentrations and also counts three times on the final map. The factor maps ‘water system per municipality’ and ‘inhabitants per water system’ count two times on the final map. The factor maps lithology, flow accumulation and slope count once on the final map.

The synthesis, called the ‘risk map’, is shown in Fig. 1. The municipality at highest risk according to this scenario is Nelas.

3.2. The ESPON methodology

The intensity of hazard was higher for the Beira Interior Norte, Dão Lafões and Pinhal Interior Norte NUTS, considering that a higher number of mines and water systems in a NUTS III corresponded to a higher intensity of hazard, as can be seen from Table 3.

Considering the same classification for population densities and GDP per capita in all of Europe, the classes presented for the ESPON methodology are explained in Ref. [2]. The GDP per capita of this Portuguese region of 10 NUTS III is all in the first class (<50). In the case of population density the

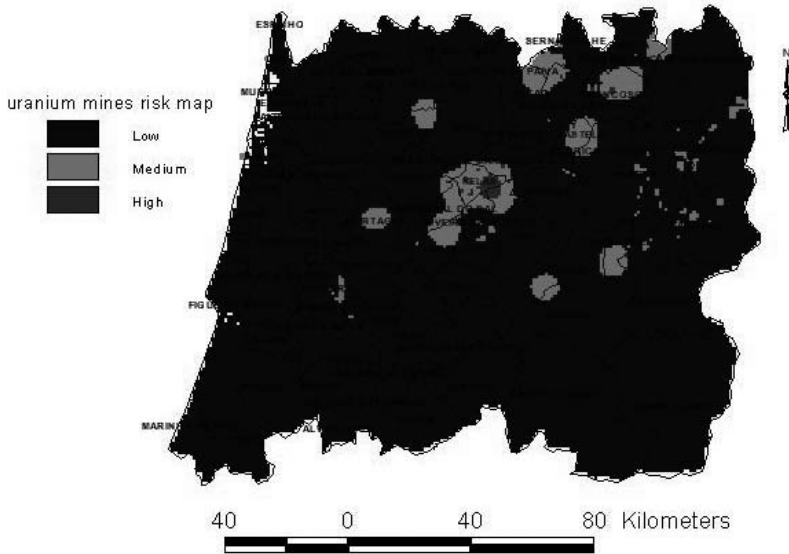


FIG. 1. Risk map of the 'uranium mining contamination' hazard in the central region of Portugal.

coastal NUTS III have class III and IV and the interior NUTS III have class II and I.

The resulting map, obtained with the intersection of Table 3 and the 'degree of vulnerability', is shown in Table 4 and represents the typology of hazards. Table 5 shows that with this methodology the NUTS III with the highest risk are Dão-Lafões and Pinhal Interior Norte.

4. CONCLUSIONS

For the first methodological approach, the final map is greatly influenced by the three weighting factors in the constituent maps. In order to evaluate the influence of each factor in the final map, the same weight should be given to all factors. Other factor maps should also be integrated into this calculation, such as natural radiation, type of storage of volumes of waste, type of ore, type of exploitation and main mineralogy of the ore body. The natural radiation could be considered a driving force indicator, and type of storage of volumes of waste a pressure indicator because it will only be considered a risk if an immediate hazard, such as a tailings dam rupture, not a long term hazard can occur. Flow

TABLE 4. TYPOLOGY OF HAZARDS [2]

Intensity of hazard	Degree of vulnerability				
	I	II	III	IV	V
I					
II	Beira Interior Sul	Pinhal Litoral Baixo Mondego	Baixo Vouga		
III	Pinhal Interior Sul	Serra da Estrela			
IV		Cova da Beira			
V	Beira Interior Norte	Dão Lafões Pinhal Interior Norte			

accumulation and slope will have to be retried because at this scale and with these classes the information obtained from these two maps is not significant. With the present risk map scenario the possible response indicators could be: ‘surveillance of the water supply systems’ and ‘conditioned water and agriculture use’ in the higher risk level.

The maps resulting from application of the ESPON methodology are very much influenced by the 50–50 weight of the ‘degree of vulnerability’ factors, population density and GDP per capita, meaning that damage to people should have a higher weight factor in the final risk assessment. Regional vulnerability factors based on inquiries from regional authorities that deal with risk issues could be considered in order to modify this weighting. Examples of regional vulnerability factors to be considered in new maps are population density, GDP per capita, number of doctors per inhabitant, population lost per NUTS III. In this case the intensity of hazard would be created with ‘type of storage of volumes of waste deposits’ as the pressure indicator and other relevant indicators.

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Paper 2

RADIOACTIVE SURVEY IN FORMER URANIUM MINING AREAS OF PORTUGAL

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Abstract

Mining of radioactive ores in Portugal started at the beginning of the 20th century and continued until 2001. About 60 sites were exploited for radium in the beginning, and later for uranium production. Radioactive ores from the majority of these mines were transported to a small number of places where physical and chemical treatment took place. There are about 3 million tonnes of radioactive spoil heaps from the processing of the uranium ore. A preliminary assessment of the radioactivity in the areas of open pit mines and milling tailings was undertaken. The results of this environmental survey are presented here. The work allowed the identification of areas with higher radioactivity and higher environmental radiation doses. The results will be of help in the designation of areas for environmental remediation work in order to ensure the radiological safety of the population.

1. INTRODUCTION

The mining of radioactive ores was started in Portugal in 1912, the Rosmaneira mine being the first mining concession recorded. From the beginning, the extraction of radium was the main objective of this mining activity and the associated uranium was not exploited, despite the fact that uranium had been known since 1841. The Portuguese mines may have produced about 100 mg of radium purified in the radium salts factory in Barração, near the city of Guarda. After 1945, when the nuclear fission chain reaction was controlled, the price of uranium rose on the international market and the production of uranium became the target of the mining activity, whereas the production of radium was discontinued.

Sixty mines were exploited for radioactive ore, most of them located in the counties of Guarda, Viseu and Coimbra (Fig. 1). A great number of them

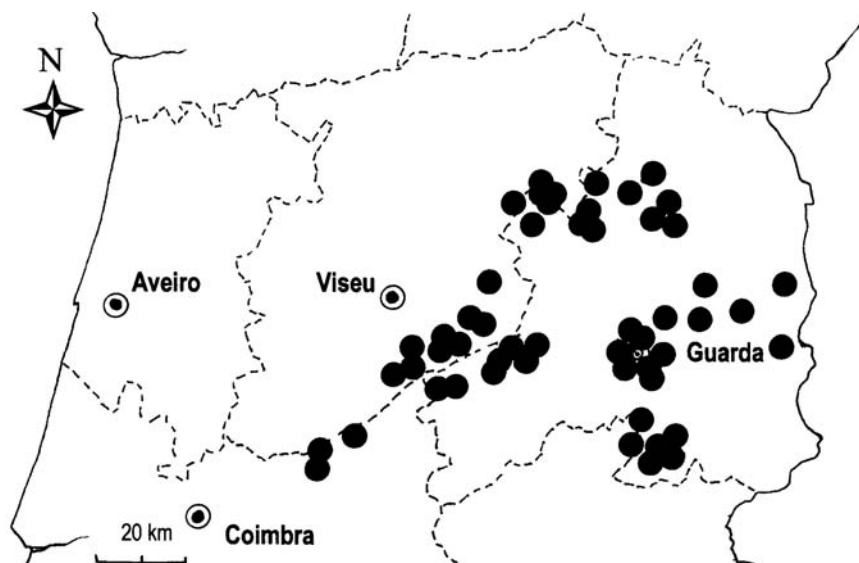


FIG. 1. Location of the main uranium mines in Portugal [1].

were of small size and exploited as open pits, although some of the larger ones were exploited as underground galleries or in a combination of both methods. In most cases the uranium ore was extracted and then transported to Urgeirica (Canas de Senhorim, Guarda) or another major mining facility for milling and extraction of the uranium, to be stocked as uranium oxide U_3O_8 . Other chemical treatment facilities were operated at Bica (Sabugal, Guarda), Quinta do Bispo and Cunha Baixa (Mangualde, Guarda), Senhora das Fontes (Pinhel, Guarda) and Castelejo (Gouveia, Guarda). The total production was about 4000 tonnes of U_3O_8 (Fig. 2).

After the cessation of mining activities only small amounts of spoil materials were left in most of the mining sites. However at the ore processing sites, particularly at those places where the ore was leached with sulphuric acid, solid waste has been stocked as tailings that still contain radioactive materials. According to Portuguese law mining companies, in this case the Empresa Nacional de Urânio (ENU SA), are responsible for the safety and security of the concessions and of wastes discharged into the environment, including acid and radioactive effluents, as well as for environmental remediation after the cessation of mining operations. However, with the end of the mining activities the company is currently in liquidation. Therefore, with the closure of a one-century cycle of uranium and radium production, there is an urgent need to assess the radioactive content of the spoil heaps and the mill tailings, as well as

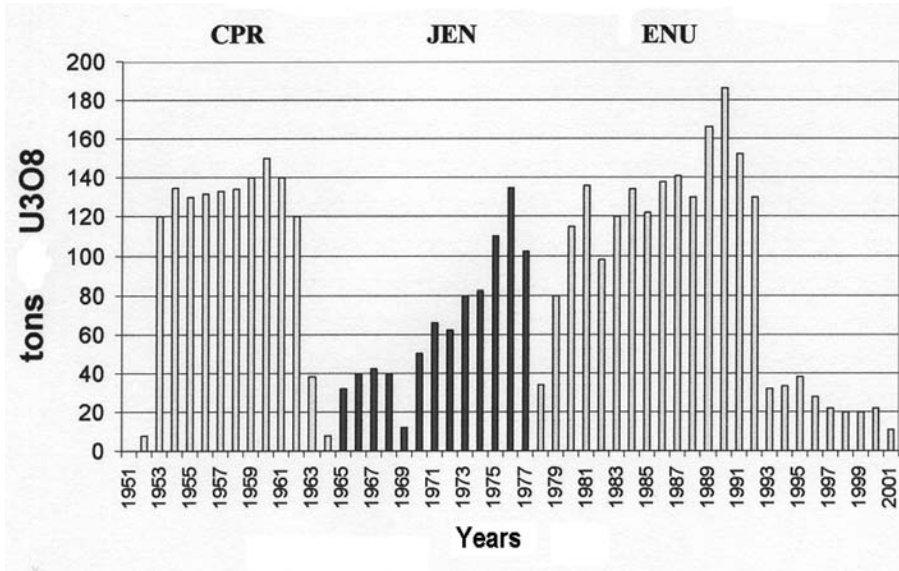


FIG. 2. Annual production of uranium concentrate by the Companhia Portuguesa do Rádio (CPR), Junta de Energia Nuclear (JEN) e Empresa Nacional de Urânio (ENU) [2].

the environmental radioactive contamination due to mining, in order to take the right steps to ensure the radiological protection of the population and of the environment [1, 2].

At the beginning of this project it was found that many of the old radioactive mines were not properly located on the maps and no longer known to the inhabitants of the region. However some sites, especially those with support facilities, have been identified, fenced in, and are under surveillance and still maintained by the mining company.

The project was planned as a preliminary survey of all the mining sites for radioactive ores in order to identify and localize them, taking precise geographic coordinates, to evaluate the environmental radiation dose rates and to collect samples of mining waste, water and soil for the determination of radionuclide concentrations in the laboratory.

2. MATERIALS AND METHODS

2.1. Identification and localization of old uranium and radium mines

This work was done between 2001 and 2003 using the information provided by the uranium mining company. Not every old mine was easily located in the field. However, by also making use of the recollections of older people in the nearby villages, it was possible to identify 58 of the old mines. Each site was referenced with a portable GPS, photographed and a topographic sketch made.

2.2. Measurement of environmental gamma radiation dose rates

The radiometric survey of the absorbed dose and dose equivalent rates was performed at each mining site using a Smart Ion portable ionization chamber (Nuclear Enterprises, UK). The environmental gamma radiation doses measured at 1 m above the ground were systematically measured using an MC-80 Mini Instruments Geiger-Müller tube. The GM tube was calibrated at the DPRSN's Metrology of Radiations Laboratory. A grid of measurement points was established at each site. In the same counties measurements were also performed at sites far from the mining areas in order to record the unmodified natural radiation background of the region.

2.3. Gamma and alpha spectrometry analysis

Samples collected in spoil heaps or geological debris from mining activity were brought to the laboratory, dried in an oven at 80°C, sieved through a stainless steel sieve with a 1 mm cut-off, homogenized and filled into 250 mL plastic cans for gamma spectrometry. The cans were kept for 35 days or more in order to allow for the attainment of radioactive equilibrium between ^{226}Ra and its radioactive daughters. Measurements were performed using Hiperpure germanium detectors (Canberra) calibrated with a radioactive standard solution QCY mix of radionuclides (Amersham), made to the same geometry as that of the samples. Gamma spectra were analysed with Genie 2000 software. A gamma spectrogram of milling waste materials is shown in Fig. 3.

Samples of mining debris and soils of the region were also analysed by alpha spectrometry. Sample aliquots were dissolved with mineral acids after addition of known amounts of radioactive tracers (^{232}U , ^{229}Th , ^{224}Ra , ^{209}Po).

Counts

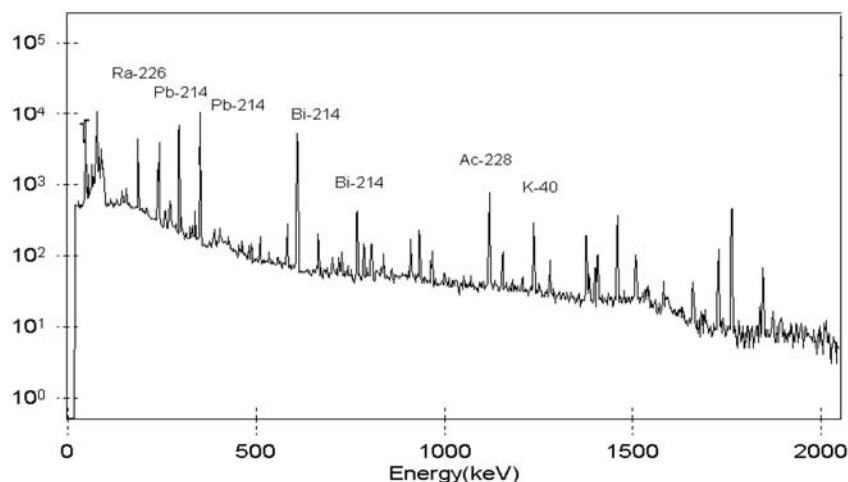


FIG. 3. Gamma spectrum of a sample from the uranium tailings of Urgeiriça.

Radiochemical separation of radionuclides was performed by radiochemistry using ion exchange chromatography columns and the purified radionuclides were electroplated onto stainless steel discs. Polonium was spontaneously plated onto a silver disc [3]. The measurements of the alpha thin sources were made with 450 mm² silicon surface barrier detectors and spectra analysed with Maestro software (Ortec-EG&G).

Water samples from underground mines and water accumulated in open pit mines were collected, filtered through 0.45 μ m pore size Millipore membrane filters, and acidified to pH<2. In the laboratory, isotopic tracers were added and radionuclides co-precipitated with MnO₂. The precipitate was redissolved and radionuclides separated, purified and electroplated for measurement by alpha spectrometry [4]. An example of an alpha spectrogram is shown in Fig. 4. The analytical quality of the results, both for gamma and alpha spectrometry, was ensured through participation in periodic exercises of inter-laboratory comparisons organized by the IAEA and the EU [5].

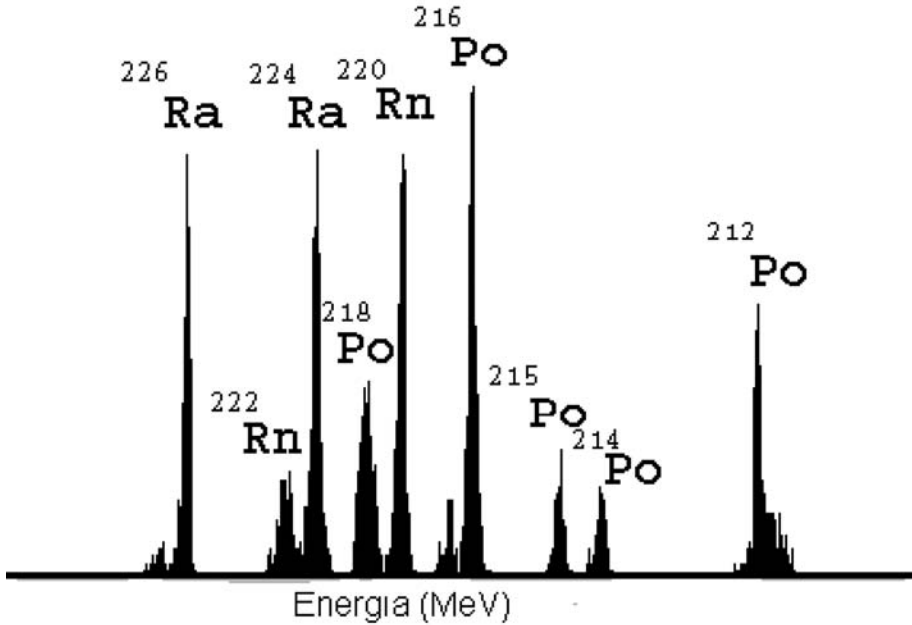


FIG. 4. Alpha spectrogram of ^{226}Ra and daughters in the water from a well near Urgeiriça.

3. RESULTS AND CONCLUSIONS

3.1. Environmental radiation doses

The results of environmental radiation dose equivalent rates measured in several places, including mining and reference areas, are given in Table 1. Comparing results for old mining sites with background sites indicates that in the majority of old mining areas the radiation dose rates are not significantly elevated above the natural background. However, in some old mines there are spoil heaps that emit gamma radiation dose rates much above the natural background.

At the mining sites where no elevated radiation dose rates were detected the most common feature on the landscape is an open pit, which indicates that the radioactive ore was almost completely extracted and transported for processing elsewhere.

Today some of these open pits are filled with water and have become seasonal or permanent ponds. Sometimes there are rock debris nearby, but in

general these are not significantly radioactive and correspond to 'sterile' overburden removed to reach the uranium ore. Some examples are the Freixinho, Fontinha and Pero do Moço mines.

At those mining sites where moderately enhanced radiation doses were measured and where open pits also exist, the spoil heaps usually correspond to rocks containing low grade uranium ore left at the site because uranium extraction was not economically profitable. These low grade ores were deposited on the surface and are subject to weathering and accessible to the public. Examples are the mines of Ribeiro do Bôco, Prado Velho and Mortórios.

At the old mining sites where ore milling and chemical processing for uranium extraction took place, fine grained sand was accumulated in tailings ponds after leaching with sulphuric acid. These waste materials still contain traces of uranium and all the progeny of ^{238}U , i.e. radium, lead, bismuth, polonium, etc. The dose equivalent rates measured on the tailings vary with the location and in some spots reach $24\ \mu\text{Sv/h}$, while the radiological background of the region typically varies between 0.2 and $0.7\ \mu\text{Sv/h}$. Examples of mining sites with such characteristics are Urgeiriça, Quinta do Bispo, Bica and Senhora das Fontes (Table 1).

Acid mine waters are to be found in some mines even though no chemical processing took place. This is the result of the use of sulphuric acid for in situ leaching of uranium from the host rock. As a consequence of this practice the acid waters in these mines contain radionuclides and heavy metals in solution. These acid mine waters constitute an environmental problem of a different nature from that posed by the uranium mill tailings. Examples of sites with such characteristics are the Cunha Baixa, Urgeiriça and Quinta do Bispo mines, where acidity is much lower than natural acidity caused by oxidation of sulphur from pyrite.

3.2. Spectrometric analyses

More than 60 samples of solid waste collected from the old mining sites were analysed by gamma spectrometry. The concentrations of gamma emitting radionuclides vary from values close to the background of naturally occurring radionuclides in soils to values much elevated due to the uranium mining activities. Table 2 shows some results of the analyses of spoil heaps and tailings from uranium mining. Taking as an example the concentrations of ^{235}U , one isotope contributing 0.7% of the mass concentration of natural uranium, activity concentrations vary from 480 to 1400 Bq/kg. However, in the same materials the activity concentrations of ^{238}U , the most abundant of uranium isotopes, contributing to 99.28% of the mass of natural uranium, vary from 3500 to 43 400 Bq/kg.

TABLE 1. RADIATION DOSE RATES MEASURED IN THE FIELD WITH AN IONIZATION CHAMBER

Mining site	Maximum (mSv/h)	Typical range (mSv/h)	Location
Reboleiro	7.0	2–4	In the village
Mortórios	3.5	1.3–3	Open pit mine
Sra Fontes	11.8	1.2–6	Agricultural field
Bica	12	3–7	Tailings/forest
Barracão	12	2–10	Village/factory
Urgeiriça	24	5–10	Tailings
Reference site			
Guarda	1.0	0.2–0.7	Near the city
Aguiar da Beira	0.5	0.2–0.5	Near the city
Belmonte	0.7	0.2–0.5	Near the city

TABLE 2. ANALYSIS OF SOLID WASTE MATERIALS BY GAMMA SPECTROMETRY: CONCENTRATION OF RADIONUCLIDES IN SAMPLES COLLECTED FROM TAILINGS (Bq/kg) (RELATIVE STANDARD ERROR <10%)

Mining site	²³⁵ U	²³⁴ Th	²²⁶ Ra	²²⁸ Ra	²¹⁰ Pb
Urgeiriça (Barragem Velha)	620	975	15 700	<98	19 700
Quinta do Bispo	480	1 490	16 700	80	27 200
Sra das Fontes	510	2 770	8 740	86	16 540
Bica	1 290	3 740	18 590	135	34 040
Vale da Arca	635	6 540	3 530	50	7 100
Barracão	1 190	400	43 440	78	24 340
Freixiosa	820	9 530	5 620	<54	5 950
Carril	1 390	33 390	18 060	115	13 390

It is interesting to compare these results with the concentrations in soils representative of the unmodified natural background (Table 3). Some of the soils analysed have been contaminated with the residues of uranium milling and the concentrations are more elevated in soils near the mines than elsewhere (Table 3). This may ease the transfer of radionuclides along the terrestrial food chain to man, starting at soil-to-plant transfer through root absorption, and accumulation of radionuclides in edible vegetables [5–8].

The water of some of the open pit mines and water from the underground mines were analysed for dissolved radionuclides by alpha spectrometry. The results shown in Table 4 show clearly that elevated concentrations of radionuclides in mine waters do exist when compared with other waters from the region. In the mine waters the concentrations of radionuclides vary with the pH (Table 4).

In the waters of open pits, with pH in general closer to neutral or slightly acidic due to the oxidation of sulphides in the rocks, the concentrations of radionuclides in solution are generally low. Nevertheless, in mines with acid waters resulting from the use of H_2SO_4 for in situ leaching, radionuclide concentrations are in general high and reach, for example, 170 Bq/L of uranium in the waters of the Quinta do Bispo mine. At the Urgeiriça mine, which has been gradually flooded by seepage of groundwaters, for years the water was pumped out and the pH adjusted by adding calcium hydroxide together with barium chloride to co-precipitate radium (Table 4). Over the last few years, the pH of the waters in the galleries was raised to about 5, but the concentrations of uranium and radium in them still remain high.

TABLE 3. ANALYSIS OF AGRICULTURAL SOILS BY GAMMA SPECTROMETRY: CONCENTRATION OF RADIONUCLIDES (Bq/kg) (RELATIVE STANDARD ERROR <10%)

Sampling site	Group	^{235}U	^{234}Th	^{226}Ra	^{228}Ra	^{210}Pb
Samples collected near old uranium mines						
Espinho	1	<6	306	260	115	310
Caldeirinhas	1	70	340	200	104	290
Quarta-Feira	1	<16	200	150	57	320
Reference areas						
Azenha	2	35	120	80	55	155
Chãos (Guarda)	2	12	168	180	80	200
Pínzio (Guarda)	2	8	150	150	55	120
Lamegal (Guarda)	2	10	190	170	170	145

TABLE 4. CONCENTRATIONS OF RADIONUCLIDES (kBq/kg) IN TAILING MATERIALS AND NEUTRALIZATION MUDS (FRACTION <63 mm) FROM PRECIPITATION OF RADIONUCLIDES FROM ACID MINE WATERS, ANALYSED BY ALPHA SPECTROMETRY

Sampling site (mine)		^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Po	^{232}Th
Cunha Baixa	Tailings	2.03	0.090	2.2	3.6	6.7	4.7	0.46
	Mud	7.4	0.31	7.3	17.8	5.1	5.3	0.30
Bica	Tailings	0.39	0.018	0.40	0.65	1.8	0.70	0.21
	Mud	10.7	0.48	11.4	30	50	29	0.18
Rosmaneira	Tailings	29	1.18	30	67	48	50	0.079

TABLE 5. CONCENTRATION OF RADIONUCLIDES IN FILTERED MINE WATERS (MBq/L) ANALYSED BY ALPHA SPECTROMETRY AND WATER pH

Mines	U_{total}	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th	pH
Bica (underground)	8 600	1 500	1 820	226	79	3.05
Cunha Baixa (underground)	4 206	84	130	247	—	3.48
Quinta Bispo (pond; in situ leaching)	169 700	1 100	1 200	87	—	2.67
Maria Dónis (pond)	100	17.2	45.1	12.6	0.26	6.70
Corga Valbom (pond)	66	12.2	30.4	17.4	0.11	5.89
Mondego Sul (pond)	4 286	353	278	18.8	0.60	5.25

In the Cunha Baixa mine, where in situ leaching with H_2SO_4 was also applied, seepage of acid waters into the shallow aquifer has occurred and they are now re-emerging in wells downstream. In some of these wells that are used for irrigation, the water pH is low and concentrations of radionuclides are high. Table 5 shows the radioanalytical results obtained by alpha spectrometry of filtered water from several mines.

The results obtained both from the field survey of mining sites and from the laboratory analyses were compared with the information available about the mining sites, which allows the grouping of sites according to the radiological risk and environmental impact observed (Table 6). This classification provides a first insight into remediation priorities and measures that may be required.

TABLE 6. PRELIMINARY RISK ASSESSMENT OF THE OLD URANIUM MINING SITES BASED MAINLY ON RADIOLOGICAL PROTECTION CRITERIA AND ASSIGNED PRIORITY FOR ENVIRONMENTAL REMEDIATION

Site containing	Priority			
	Very high	High	Moderate	Low
Radioactive tailings/mud	Yes	Yes	No	No
Environment radiation dose	>1 mSv/a	>1 mSv/a	Low	Background
Acid waters	Yes	No	No	No
Contaminated facilities	May exist	No	No	No
Other problems (safety, pond water use...)	May exist	May exist	Yes	Yes
Number of sites estimated	5	3–5	20	30

4. CONCLUSIONS

This preliminary survey of all the radium and uranium mining and milling activities in Portugal allowed the precise localization of old mines and the identification of spoil heaps resulting from the mining, including those that are still under institutional control and monitored, as well as those already forgotten. The results obtained lead to the conclusion that many of the small mines that were exploited as open pits are not a source of significant exposure to ionizing radiation for the public. However, these old mines may still pose some non-radiological risks such as those related to the physical safety of the sites (shafts, pits), to the potential use of waters for irrigation, to the potential use of sand and gravel as construction materials, etc. These risks should not be overlooked or forgotten and, although they are not radiological in nature, they need to be taken into account in the environmental remediation plans for these sites.

In contrast to these sites, the old uranium and radium mines and ore processing facilities with tailings containing radioactive materials represent a source of radiation exposure to the population and may represent a significant radiological risk to the public. Fortunately, these sites are few in number, though the amount of radioactive waste accumulated is estimated to be as high as 3 million tonnes. Furthermore, some mines do have acid waters containing elevated concentrations of radionuclides.

The findings of this survey underscore the need for corrective measures in the old uranium mines. These measures need to be adequate to deal with the environmental and waste problems existing at each site. In the phasing out of the uranium mining activities and decommissioning of associated facilities in the country, the recommendations of the international organizations and safety standards for radiological protection of humankind and of the environment need to be duly taken into account [9].

ACKNOWLEDGEMENTS

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Paper 3

RADIOACTIVITY IN PUBLIC WATER SUPPLIES IN THE URANIUM MINING REGIONS OF PORTUGAL

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Abstract

Most of the old uranium mines in Portugal are located in the granite regions of the centre-north of Portugal (Beiras), in regions with relatively high population density. Most of the public water supplies for the villages and cities are from groundwater or from surface water reservoirs constructed on the rivers of the region. A survey of the radioactivity in the water supplies was carried out in order to investigate the concentration of radionuclides from the uranium family such as uranium, radium, polonium and lead radioisotopes. Radon was also determined, as well as total alpha and total beta radioactivity, in order to assess the human exposure through consumption of drinking water. In general the radioactivity content of waters is reasonably low, not too high for human consumption. In several cases, however, the concentrations of radionuclides such as dissolved radon, polonium and radium, or even uranium, are elevated with total alpha concentrations above 0.1 Bq/L. Cases of public water supplies with very high concentrations of dissolved radionuclides are rare and in general apply to spring waters that may come into contact with rocks containing localized mineralizations of uranium. The highest concentrations of radionuclides were measured in water from wells in areas under the influence of old mines, especially the ones where in situ leaching with sulphuric acid had been performed in the past. This is the case, for example, for Cunha Baixa and Quinta do Bispo, two old mines draining into the same valley. At present, most of these wells are used for irrigation purposes and drinking water comes from other supply sources. Consideration is given to the radiation doses to members of the public through water consumption from public supplies of these regions.

1. INTRODUCTION

The uranium mining activities of Portugal were concentrated in the centre-north regions of the country, in the granite massif of the Iberian Meseta

and at its edge. About 60 deposits of uranium were exploited during the 20th century, with different degrees of environmental impact according to the type of exploitation and amount of radioactive ore extracted. A preliminary survey of the radioactivity at old mining sites provided evidence of enhanced concentrations of radionuclides in the environment in some locations. Especially in locations where sulphuric acid was applied for in situ leaching and around chemical facilities for uranium ore processing, the dispersal of radionuclides in the environment appears to be wider.

In the centre-north region there are several main cities and hundreds of small towns with a total population of about 3–4 million inhabitants. In the region, the water supplies are of several types. Reservoirs (dams) on major rivers are used as water supply for large cities, although private wells and spring sources are used as water supplies in small communities.

As rivers in the region flow through the counties where uranium mining activities took place in the past and most of the bedrock is either granite or schist, there is a potential for the presence of noticeable radioactivity in water. This may occur either from natural geochemical processes involving, for example, dissolution of radioelements from the bedrock, or from anthropogenic activities enhancing concentrations of naturally occurring radionuclides.

The objectives of this study were to investigate the radioactivity in the public water supplies and determine the concentrations of the main radionuclides present, especially alpha emitters, and to assess the radiological risk for populations that use the current water supplies. Furthermore, this study was to help identify water supplies, public or private, not suitable for human consumption, and assist in clarifying the relationship between radioactivity in water and past uranium mining activities and mining residues.

2. MATERIALS AND METHODS

Water samples were collected in the centre-north region from public water supplies at the tap, from deep wells (artesian boreholes), from reservoirs and rivers, and from private superficial wells and spring waters used to feed public fountains in villages. Large volume water samples were collected directly into 10–35 litre capacity polyethylene drums, acid pre-washed, and washed three times with small volumes of the same water as used for the analyses. Water samples were filtered the same day, in the region, through 142 mm diameter membrane filters with an $0.45\ \mu\text{m}$ pore size, using pressure from a peristaltic pump. The filtered water was collected and acidified with HNO_3 to a pH of around 1–1.5 and saved for analysis in the laboratory. Filters with the particulate matter were retained for analysing radionuclides

associated with particles. A volume of the same water was filtered through a pre-weighted smaller filter for determination of the dry weight concentration of suspended particles.

In the laboratory, isotopic tracers for the radioelement of interest, i.e. ^{232}U , ^{229}Th , ^{224}Ra , ^{209}Po and stable Pb carrier were added to the filtered water samples that were then thoroughly homogenized by prolonged air bubbling and co-precipitated with MnO_2 . The precipitate was collected, re-dissolved and the radioelements separated and purified by using ion exchange resins in chromatography columns (Fig. 1). The purified radioelements were then electroplated on stainless steel discs as thin sources. Polonium was spontaneously plated onto a silver disc [1–3]. Filters containing the suspended matter were dissolved with mineral acids and the same procedure of separation and purification was applied in order to prepare thin sources on metal discs suitable for alpha spectrometry.

Measurements of alpha emitting radionuclides were made with ion-implanted silicon surface barrier detectors with active surfaces of 450 mm^2 using an Octete alpha spectrometer (Ortec-EG&G), and the spectra analysed with the Maestro software. The detectors were energy calibrated with a certified electroplated Pu–Am–Cm alpha source from Amersham (UK). The use of isotopic tracers added to the samples as the internal standard allows for individual determination of the radiochemical yield. The precision of the analytical methods was repeatedly tested through analyses of certified reference materials and by participation in intercomparison exercises organized by the IAEA.

Determination of radon (^{222}Rn) dissolved in water was done in separate samples collected in the field for this purpose. As the measurement was made by liquid scintillation counting (LSC), 10 mL were drawn with a syringe directly into a low background glass vial containing 10 mL of scintillation cocktail (OptiFluor-O, Packard). The vials were counted with a LSC (Tri-Carb 3170 TR/SL, Packard), usually for periods of 60 minutes. The procedure and calibration method are described in more detail in Ref. [4]. Total alpha and total beta radioactivity in the water were determined in the same samples using 2 litres of water. These samples were concentrated in Pyrex glass beakers by gentle heating to a volume of 50 mL and salts were precipitated by the addition of 1 mL of concentrated sulphuric acid. Sulphate residues were calcinated for one hour at 350°C and then transferred to a stainless steel planchet of 50 mm diameter. Alpha and beta activities were measured in the same planchet in a proportional gas flow counter (Canberra XLB, Tennelec Series 5), previously calibrated against ^{241}Am and ^{40}K standard sources for alpha and beta activity, respectively [5–6]. The activity concentrations reported were determined with accuracies better than 5% of relative standard error.

Water sample +
HNO₃ + Isotopic tracers

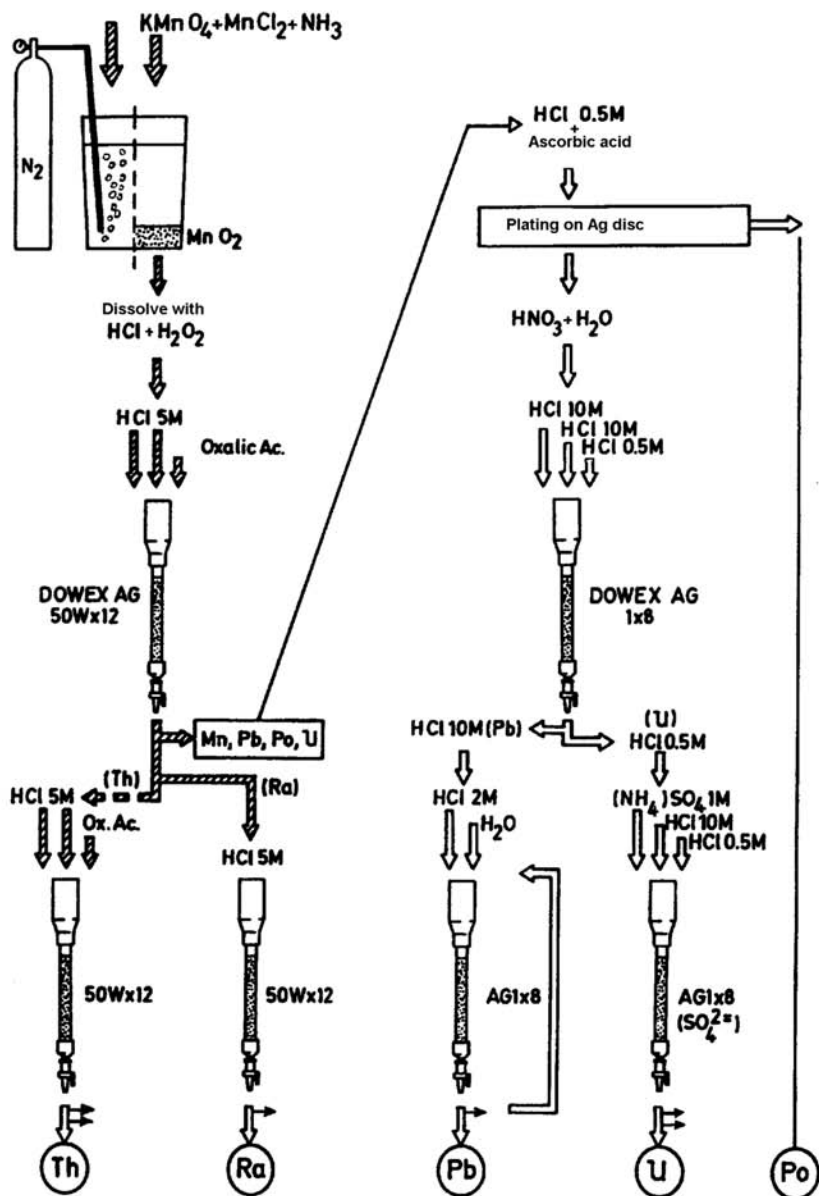


FIG. 1. Flow diagram of the sequential radiochemical separation of natural radionuclides from a water sample for electroplating and determination by alpha spectrometry [1].

3. RESULTS AND DISCUSSION

The results of the analyses of public water supplies (tap waters) in several towns and cities in the uranium region are shown in Table 1. Activity concentrations of particle reactive radionuclides such as ^{232}Th and ^{210}Po are very low. Concentrations of more water soluble elements such as uranium, radium, and especially ^{210}Pb are higher. Therefore, in the public water supplies of these cities the main alpha emitters are radionuclides from the natural uranium series (^{238}U), including the uranium isotopes themselves. The beta emitter ^{210}Pb is also a major contributor but its granddaughter ^{210}Po , an alpha emitter, is present in concentrations lower than those of ^{210}Pb , probably due to its removal from solution by adsorption onto the suspended particles and pipe walls [2].

The analyses of a large collection of tap water samples from the region for total alpha and total beta radioactivity show concentrations generally below the maximum recommended limits applicable to water for human consumption, i.e. 0.1 Bq/L and 1 Bq/L for alpha and beta activity, respectively, as set in EU Directive 98/83 CE [7] (Fig. 2).

Waters from deep wells were analysed by alpha spectrometry and results are shown in Table 2. Activity concentrations of radionuclides are higher than in tap waters. Particularly ^{210}Po , ^{210}Pb and ^{226}Ra attained very high concentrations. In one case uranium and ^{210}Po are present in exceptionally high concentrations. One can notice that ^{232}Th is, however, present only in low concentrations. It is likely that these waters from deep aquifers have dissolved radionuclides from host minerals present in the granites.

TABLE 1. CONCENTRATION (MBq/L) OF ALPHA RADIONUCLIDES IN WATER FROM PUBLIC SUPPLIES (TAP WATER, NOT FILTERED) OF SEVERAL CITIES IN THE CENTRE-NORTH OF PORTUGAL

	^{238}U	^{235}U	^{234}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Sabugal	13.6	0.59	12.9	0.64	38	1.0	0.16
Belmonte	6.1	0.28	6.0	3.7	24	2.0	0.17
Quartafeira*	42.1	1.8	35.4	10.6	100	7.5	—
Guarda	1.6	0.08	2.0	3.2	11	2.7	—
Trancoso	12.2	0.56	13.6	9.8	39	3.2	—
Mangualde	20.2	1.0	18.4	12.8	48	3.6	0.14

* Water from a spring source in a nearby mountain.

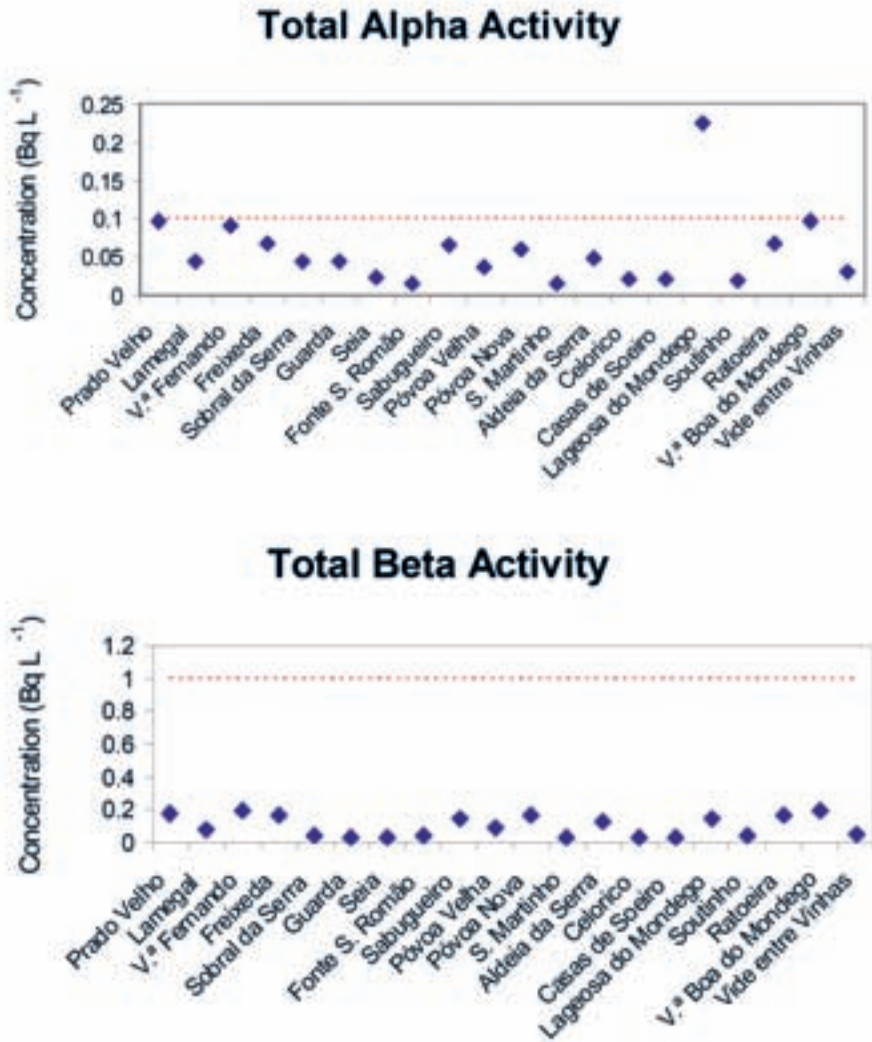


FIG. 2. Total radioactivity in water from public supplies (tap water, not filtered) of cities and villages of the centre-north of Portugal. Dotted lines indicate maximum recommended concentrations acceptable in waters for human consumption.

Surface waters from rivers and reservoirs contain low concentrations of dissolved radionuclides (Table 3). The concentrations in these waters can be compared with concentrations in waters sampled from the tap (Table 1), keeping in mind that surface waters were filtered before analysis and tap waters were not. Waters from these rivers and reservoirs are the source of

public water supply networks for towns and cities in this region. These waters are treated to reduce suspended particles, organic matter and bacteria, but particle removal is not as effective as filtration through membrane filters. Nevertheless, concentrations of uranium series radionuclides and thorium in the soluble phase are generally low in these surface waters as well, and are in reasonable agreement with the concentrations measured in tap water samples.

Radon concentrations in waters from various sources display values exceeding 1000 Bq/L. The highest values were measured in waters from fountains supplied by spring waters in villages and in waters from deep wells (Fig. 3). Most of the tap waters, more than 50%, contain dissolved radon in concentrations of less than 100 Bq/L, whereas more than 50% of waters from deep wells display concentrations above 100 Bq/L. Water from near surface wells generally also display low concentrations of radon comparable to those in tap waters.

TABLE 2. CONCENTRATION OF RADIONUCLIDES (MBq/L) IN UNFILTERED WATER FROM ARTESIAN DEEP WELLS USED FOR HUMAN CONSUMPTION

	^{238}U	^{235}U	^{234}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Outeiro	130	5.5	181	223	802	3700	—
Vila Novinha	52.9	2.1	57.4	9	—	7.6	0.12
Benvende	1640	68	1760	757	—	1540	4.6
Pisões	48.7	2.3	55.2	31	—	188	0.48
Vale de Madeiros	488	20.8	520	122	—	34	0.53

TABLE 3. CONCENTRATIONS OF RADIONUCLIDES (MBq/L) IN THE DISSOLVED PHASE OF WATER FROM RIVERS AND SURFACE RESERVOIRS (FILTERED SAMPLES)

	^{238}U	^{235}U	^{234}U	^{226}Ra	^{210}Po	^{232}Th
River Alva	3.4	0.17	3.3	13	10	0.13
River Seia	2.5	0.10	2.6	9	7	0.28
River Mondego	2.6	0.12	3.1	4	6	0.20
River Távora	8.0	0.44	8.8	21	15	0.30
Teja reservoir	60.6	2.9	61.1	12	16	0.20
Aguieira Reservoir	3.6	0.2	3.5	2.1	—	—

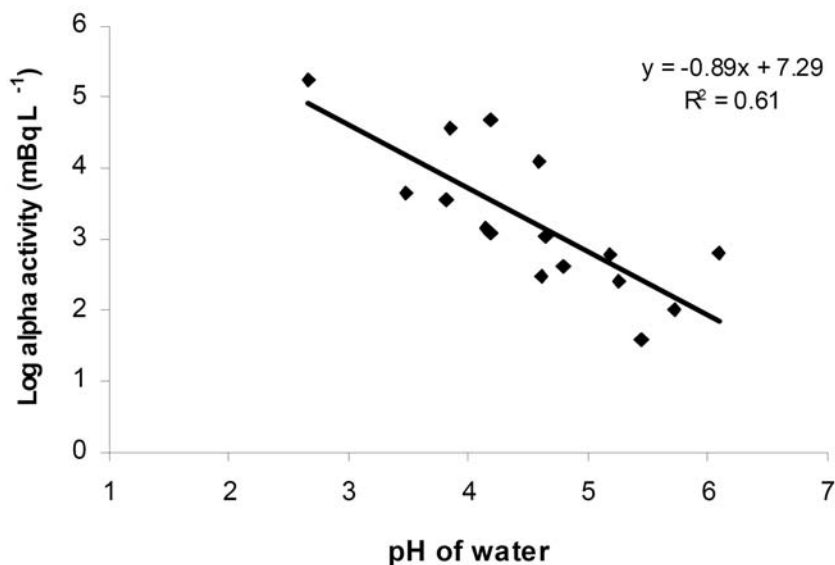


FIG. 3. Total alpha radioactivity and pH of water in wells in the area of Cunha Baixa village. The straight line is the best fit to experimental points. The correlation is statistically significant at $p < 0.001$.

Waters from shallow wells with water levels 1–3 m below the ground in the area of the village of Cunha Baixa have been analysed by alpha spectrometry. This village was the site of an underground uranium mine which was exploited until a few years ago, where the in situ acid leaching technique was applied. The results for the filtered water from wells are shown in Table 4 and indicate moderate concentrations of uranium series radionuclides in solution. However, these concentrations are higher than concentrations measured in the rivers of the region (Table 3) and suggest an enhancement of the natural radionuclide concentrations. The water from these wells is also acid, with high concentrations of SO_4^{2-} ions. The plot of total alpha activity against pH confirms a statistically significant correlation between both parameters (Fig. 3). Water from wells at greater distances (>1000 m) from the Cunha Baixa mine contain lower concentrations of radionuclides, typically one or two orders of magnitude less than those in Table 3, highlighting the contamination of the shallow aquifer with acid mine waters in the vicinity of Cunha Baixa.

Seepage from acid mine waters and surface runoff from the tailings may impact the waters in the creeks and groundwaters in the shallowest aquifer. Measurements made in well waters in the area of Cunha Baixa and Quinta do

TABLE 4. CONCENTRATION OF RADIONUCLIDES (MBq/L) IN FILTERED WATER FROM SUPERFICIAL WELLS (PRIVATE SUPPLIES) IN THE AREA OF CUNHA BAIXA

Well no.	^{238}U	^{235}U	^{234}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
13	42.8	1.8	37.0	13	—	7.8	—
14	11.0	0.5	10.5	12	—	4.4	—
16	123	5.4	120	104	10	72	—
18	256	10.8	243	45	27	21	—
19	74.7	3.7	74.7	47	32	32	—

Bispo, two major uranium mines, show low pH and higher than normal radionuclide concentrations (Table 3). The flow of surface waters and the trends in concentrations do indicate intrusion of acid and radioactive mine water in the waters from wells used for irrigation in this area. This contamination of the shallow aquifer is measurable today at a distance of about 300–500 m from the mine, but the rate of migration of the contamination is not known.

The regular ingestion of water from public supply networks of the region does not pose a radiological hazard to the population. The dose received from the radionuclides ingested with this water will not exceed the annual indicative dose of 0.1 mSv/a [7]. However, some small communities consuming water from deep wells and fountains fed by spring waters may be exposed to radiation doses exceeding the recommended limit due to relatively high concentrations of radon, radium and polonium dissolved in water, all naturally occurring.

Surface waters and aquifers under the influence of acid and radioactive mine waters are contaminated in the vicinity of the Cunha Baixa and Quinta do Bispo uranium mines. A similar situation may exist in other mining areas. These waters are not suitable for human consumption both because of their being acid and because of the dissolved radioactive elements. Consumption of this water would expose the inhabitants to radiation doses above the recommended dose limits. Furthermore, the actual use of these waters for irrigation purposes may facilitate the food chain transfer of radionuclides and requires adequate monitoring, as discussed previously and recommended [8–10].

4. CONCLUSIONS

The analytical results of waters collected at the point available to consumers, i.e. tap water from public water supply networks, indicated that in most towns and villages their consumption does not pose a radiological hazard. Concentrations of total alpha and total beta radioactivity are, indeed, generally below the recommended maximum concentrations. Analysis of individual radionuclides both in tap water (as available to the consumer) and surface water from reservoirs and rivers, filtered through 0.45 μm filters, show that concentrations of uranium, radium, lead and polonium, as well as dissolved radon, are generally low. Because most of the public water supply networks to cities and towns use these surface reservoirs, the radioactivity concentrations and radiation exposure are consequently low.

Waters from deep wells may contain relatively high concentrations of several alpha emitters such as uranium, radium, lead and polonium, probably dissolved from the bedrock of the aquifer. Generally, these deep waters also display the highest concentrations of dissolved radon. There is no clear relationship of these radionuclide concentrations to uranium mining and it is likely that the elevated radionuclide concentrations are entirely of natural origin. Regular consumers of some of these waters are likely to receive radiation doses higher than consumers of the water from public supply networks.

Water from creeks receiving drainage from uranium mining and milling tailings contains enhanced concentrations of radionuclides both in solution and in the suspended matter. Waters from wells in the surroundings of old uranium mines, especially those where sulphuric acid was used, contain elevated concentrations of radionuclides and it was shown that a correlation exists between water pH and radioactivity. These wells draw water from the shallow aquifers. The consumption of this water could pose a radiological risk to humans through ingestion. Direct consumption of water does not take place, but these waters are used for irrigation of horticultural produce and are the pathway for contamination of soils and radionuclide accumulation by plants.

The dispersion of radionuclides via mine drainage by surface runoff from tailings and seepage of acid groundwaters seems to have a short range, at least at Cunha Baixa. However, the circulation of contaminated waters in the shallow aquifers and the circulation in the deep aquifer have not been investigated yet. Because waters from the deep aquifer in the region already have high radionuclide concentrations due to natural geochemical processes, the possible enhancement of radioactivity due to uranium mining should be carefully investigated before remedial actions are undertaken.

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Paper 4

APPLICATION OF RISK BASED COST–BENEFIT ANALYSES FOR DECISION MAKING IN ENVIRONMENTAL RESTORATION OF URANIUM MINING AND MILLING SITES

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Abstract

Active and abandoned uranium mining and milling sites can represent complex environmental situations, where health risks and environmental detriments may result from radon exhalation and dispersion of radioactive dust from mine wastes as well as from the discharge of contaminated mine waters into surface and groundwaters. The paper outlines a methodology for the evaluation of remediation measures in the framework of the closeout of uranium mining and milling facilities in eastern Germany. The decision making process for the remediation of large waste rock dumps (total volume approximately 125 million m³) at a uranium mining site is used as an example. In the context of this approach, appropriate and sustainable remediation measures should (1) reduce the environmental impacts from the waste rock dumps to acceptable levels, and (2) have the best cost–benefit ratio (i.e. the ‘lowest overall costs’). These ‘overall costs’ comprise direct short term costs for remediation measures (e.g. application of engineered covers, backfill of mine waste into open pits), long term costs (e.g. for monitoring, maintenance, seepage collection/treatment), as well as the monetary equivalents of remaining risks for human life and health and impacts on the environment — evaluated by geochemical modelling/air dispersion modelling — in the long term. Uncertainties in the costs and benefits of the remediation measures are addressed by stochastic methods (Monte Carlo simulations).

1. INTRODUCTION

The cleanup of contaminated sites requires appropriate and efficient methodologies for decision making about priorities and the extent of remedial measures that aim at two usually conflicting goals: to protect people and the environment by reducing detrimental impacts to the extent feasible, and to minimize the expenditure of money and other resources. Extensive mining of uranium ores, polymetallic sulphide ores, or coal and lignite often requires considerable cleanup efforts to protect the environment and the public. This

can be seen for example from various projects in the context of the UMTRA (Uranium Mill Tailings Remedial Action) and CERCLA (Comprehensive Environmental Response, Compensation, and Liability Act; commonly known as 'Superfund') programmes in the United States of America [1] or the estimated financial liabilities associated with mine tailings and waste rock in Canada [2].

Active and abandoned uranium mining and milling sites can represent complex environmental situations, where health risks and environmental detriments may result from radon exhalation and dispersion of radioactive dust from mine wastes, as well as from the discharge of contaminated mine waters into ground and surface waters. Furthermore, particularly at abandoned mining sites, other mining related hazards (e.g. slope stability of waste dumps, failure of tailings dams, etc.) have to be taken into account.

This paper outlines the methodology employed for the evaluation and selection of cost effective and sustainable remediation measures in the framework of the closeout of uranium mining and milling facilities in eastern Germany. The decision making process for the remediation of the waste rock dumps at a large uranium mining site is used as an example, emphasizing the use and the significance of predictive geochemical models to assess the contaminant discharge into surface and groundwaters.

2. SITE DESCRIPTION

The former German Democratic Republic (GDR) was one of the largest uranium producers in the world. Its cumulative uranium production of approximately 220 000 t U between 1945 and 1990 was exceeded only by the USA and Canada. The closeout and remediation of the various mining and milling sites of the former Soviet–German mining company WISMUT, which were often situated in rather densely populated areas, is one of the largest remediation projects in the world at present with an overall budget estimate of about €6.6 billion.

The mining site near Ronneburg in Thuringia was the most extensive uranium mining site in eastern Germany. Between 1952 and 1990 approximately 125 million m³ of uranium ore were produced from underground and surface mines. Uranium production from this site (approximately 113 000 t) contributed about 50% of the total uranium production of the GDR. The uranium ores are hosted predominantly in Paleozoic (black) shales, carbonate rocks and basic volcanic rocks (diabase) and consist mainly of pitchblende and coffinite. The uranium ore is accompanied by sulphide mineralization (mainly pyrite) and minor cobalt and nickel arsenides. The mine workings comprised

several (connected) underground mines that covered an area of more than 50 km², and an open pit that had a volume of approximately 160 million m³. During the lifetime of the mines, about 125 million m³ of waste rock from underground and surface mining were placed on various dumps that ranged in size between some 100 000 m³ and up to 65 million m³, covering an area of approximately 4.7 km² (Table 1). In addition, some 80 million m³ of waste rock were placed in the mined-out part of the open pit before mining activity ceased. Environmental detriments and health risks associated with these waste rock dumps comprise, in particular the exhalation of radon; the dispersion of radioactively contaminated dust; and the discharge of acid and neutral seepage with high levels of uranium, heavy metals and salts into surface and groundwater bodies (Table 2).

TABLE 1. SIZE AND SEEPAGE CHARACTERISTICS OF WASTE ROCK DUMPS AT THE RONNEBURG URANIUM MINING SITE BEFORE THE BEGINNING OF REMEDIATION MEASURES

Heap	Volume (million m ³)	Area (×10 000 m ²)	Seepage characteristics
Absetzerhalde	65.8	224.7	Acidic
Innenkippe ^a	64.0	—	Acidic/neutral
Nordhalde	27.2	83.9	Acidic
Heap Paitzdorf	7.6	24.9	Neutral ^b
Gessenhalde ^c	6.9	28.7	Acidic
Heap Reust	6.3	20.5	Neutral
Heap Beerwalde	4.5	23.9	Neutral ^b
Heap Drosen	3.5	22.8	Neutral
Heap 4	0.9	7.8	— ^d
Heap 370	0.8	6.0	Neutral
Heap Korbussen	0.4	4.1	Neutral
Heap 377	0.3	2.9	Neutral/slightly acidic
Heap 381	0.1	1.0	— ^d
Diabashalde	0.1	1.0	Neutral

Notes: ^a Waste rock placed in the open pit during production.

^b Neutral seepage but very high sulphate discharge, indicating high sulphide oxidation rates.

^c Acid leach heap.

^d No seepage observed.

TABLE 2. SEEPAGE QUALITY OF SELECTED WASTE ROCK DUMPS AT THE RONNEBURG MINING SITE

Parameter	Unit	Nordhalde	Heap Beerwalde
pH	(—)	2.0–3.5	7.5–8.0
Ca	(mg/L)	200–650	270–430
Mg	(mg/L)	800–2 800	4 200–8 000
SO ₄	(mg/L)	5 000–20 000	19 000–29 000
HCO ₃	(mg/L)	<1	200–600
Fe	(mg/L)	300–3 200	0.1–2.0
Al	(mg/L)	200–400	0.1–1.5
Mn	(mg/L)	20–180	2.5–22.0
U	(mg/L)	0.4–6.5	3.5–5.5
²²⁶ Ra	(mBq/L)	10–200	10–300
Ni	(mg/L)	24.0–38.5	0.1–1.2
Co	(mg/L)	6.5–13.5	0.02–0.25
Cu	(mg/L)	2.5–5.0	0.02–0.05
Cd	(mg/L)	0.2–0.5	<0.05
Zn	(mg/L)	16.5–26.0	0.1–0.25

Key issues of the decommissioning of the Ronneburg mining site comprise: the flooding of the extensive underground mining system; the remediation of the open pit; and the remediation of the waste rock dumps. Further measures include the demolition of radioactively contaminated buildings, as well as the construction of a water treatment plant. Potential remediation measures for the waste rock dumps include in situ remediation using various types of engineered covers or the relocation of mine waste into the open pit and cover placement over the backfilled pit. The backfilling of parts of the open pit and the use of a cover is necessary in any case to enhance the slope stability of the pit and to reduce radiological hazards resulting particularly from radon exhalation. Even after remediation, the waste rock dumps and the backfilled open pit, which is hydraulically connected to the underground mining system, will act as long term sources of contaminants, having potentially detrimental effects on surface and groundwater quality.

3. ASSESSMENT OF REMEDIATION OPTIONS

3.1. Methodology

The cleanup of contaminated uranium mining and milling sites must take into account a variety of different contaminants and radiological and non-radiological risks to humans and the environment arising for various exposure pathways. In general, the following factors have to be included in the site assessment and the evaluation of remediation options:

- (a) Health damage due to radiological risks (external irradiation; incorporation of radioactive substances), or through carcinogenic and/or toxic substances (e.g. arsenic, heavy metals);
- (b) Environmental detriments such as damage to ecosystems such as lakes and rivers due to the discharge by contaminated mine waters;
- (c) Damage to resources due to contamination of groundwater and surface water bodies;
- (d) Direct physical risks (e.g. the danger of dam failures) and risks ensuing from the cleanup activities themselves (e.g. work or traffic accidents).

To determine a cost effective balance between the reduction of risks and the required financial efforts for the reclamation work at the eastern German uranium mining and milling sites, an integrated assessment method has been developed. This approach is based upon a quantification of risks to humans and ecosystems caused by radiological and chemical contaminants, and is described in detail in Refs [3, 4].

A key aspect of this approach is the monetarization of risks and damages, thus making them comparable to the financial efforts required for mine site reclamation (see Fig. 1). Human health risks are expressed as a ‘mean effective loss of life expectancy’, which is converted into an equivalent monetary value as the basis for the cost–benefit analysis. Damage to ecosystems, in particular surface water systems, and resources is similarly expressed in monetary terms, based on the societal willingness and ability to pay for the prevention or mitigation of ecological damage.

In the context of the developed methodology, appropriate and sustainable remediation measures for the uranium mining sites should reduce the discharge of aquatic contaminants and the dispersal of radon and radioactive dust from the sites to acceptable levels (i.e. below existing German guidelines, to radiation doses of less than 1 mSv/a for the general public in the long term), and should provide the best cost–benefit ratio (i.e. the ‘lowest overall costs’). These ‘overall costs’ comprise the direct short term costs for

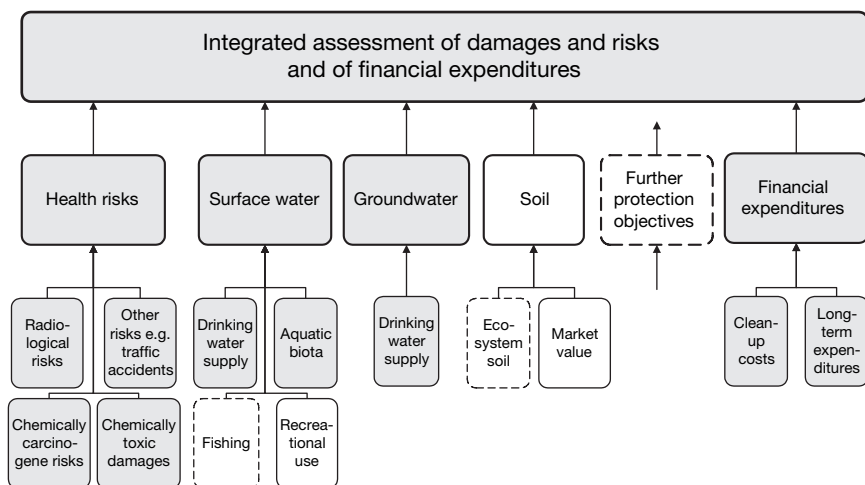


FIG. 1. Elements of the integrated assessment approach.

remediation measures (e.g. application of engineered covers on mine wastes; backfilling of mine waste in underground workings or open pits); long term costs (e.g. costs for monitoring, maintenance, seepage collection and — if required — water treatment) as well as the monetary equivalents of the remaining risks for human life and health and impacts on the environment. Cleanup options can be assessed in terms of their benefits in relation to the effort (i.e. financial expenditures) required and a basis for rational and transparent decisions about cleanup measures is established. Uncertainties in the basic data and assumptions made are taken into account using probabilistic simulation techniques (Monte Carlo simulations).

The regulatory authorities dealing with radiation protection during the remediation of uranium mining and milling sites in the former GDR require that a time frame of at least 200 years be considered in the evaluation of potential remediation measures. Decisions regarding cost effective as well as sustainable reclamation measures thus require an assessment of the long term effects of the remediation measures and their permanence, as well as an assessment of the associated long term costs.

4. PREDICTIVE MODELLING

One major issue at many mining sites with respect to environmental impacts in the long term is a continuing discharge of contaminated mine waters.

Sustainable measures for the closeout of a mining operation or the remediation of abandoned mining sites have to take into account that mine wastes, open pits and underground workings may serve as significant sources of aquatic pollutants for several hundred years and drastic changes in mine water quality may occur at some point in the future [5]. Therefore, in the framework of the decision making process, reliable predictions of the future quality of seepage water or mine water are indispensable to assess water treatment costs, radiological and toxic risks resulting from the use of surface and groundwaters, and associated environmental detriments (e.g. regarding aquatic biota).

In this context we used extensive hydrogeological and geochemical modelling to evaluate the consequences of various remediation options for the waste rock dumps at the Ronneburg uranium mining site (e.g. collect and treat option; application of engineered covers to limit the oxygen ingress and reduce infiltration and radon exhalation; backfill into the open pit). This is described in more detail in Ref. [6]. The models employed for these predictions take into account particularly:

- (a) Chemical reactions controlled by kinetics or limited by mass transfer constraints (e.g. acid generation due to chemical and microbiological sulphide oxidation, buffering reactions, chemical weathering);
- (b) The formation/dissolution of secondary minerals;
- (c) Retardation processes (e.g. sorption, co-precipitation). They allow long term predictions of seepage and groundwater quality. Furthermore, the convective and diffusive transport of oxygen in the waste dumps, which is an essential consideration for calculation of sulphide oxidation rates, heat and vapour transport due to the exothermal sulphide oxidation, and changes in particle size due to physical and chemical weathering processes are addressed.

In addition to sensitivity analyses for some important input parameters (e.g. infiltration rates, oxygen diffusion rates, availability of sulphide and carbonate minerals), probabilistic Monte Carlo simulations, using probability distributions for various input parameters, were performed to address and keep track of uncertainties in the input parameters for the geochemical modelling.

The results of the detailed simulations of the discharge of various contaminants (e.g. radionuclides, heavy metals, salts, acidity) from the mine wastes were combined with the results of the hydrogeological and geochemical modelling of the flooding of the underground mine [7] and served as an integral part of a comprehensive assessment of the effects of the site reclamation on the long term quality of surface and groundwaters in the Ronneburg area.

5. SUMMARY AND CONCLUSIONS

Taking into account the results of the air dispersion modelling for the dispersion of radon and radioactive dust for the various reclamation options considered, the following conclusions for the remediation of the waste rock dumps at the Ronneburg mining site were derived from the integrated assessment of costs and benefits of remediation measures:

- (a) In situ remediation of waste rock dumps employing engineered covers comprising a water saturated clay liner to minimize oxygen ingress (and thus reduce the sulphide oxidation rates) and radon exhalation will prevent waste rock dumps that are currently neutral from turning acidic in the future;
- (b) The relocation of waste rock material from the dumps with the highest contamination potential, i.e. the acid leached Gessenhalde (6.5 million m³), the Absetzerhalde (65 million m³), and the Nordhalde (27 million m³) into the open pit is the remediation option associated with the lowest long term detrimental impacts on surface and groundwaters and with the lowest overall costs;
- (c) The relocation of other, currently not acid generating waste rocks into the open pit will not increase the cumulative contaminant discharge from the backfilled pit into the environment within the next centuries.

Technical measures during backfilling of the pit which are recommended to minimize the long term contaminant discharge from the backfilled pit include:

- (1) The placement of acidic and potentially acid generating mine wastes below the future (i.e. after flooding of the underground mine) water table;
- (2) Addition of lime to acidic material;
- (3) Compaction of thin layers of waste rock during backfilling to reduce hydraulic conductivity, oxygen ingress and radon exhalation, as well as to avoid cover failures in the future due to settling.

The recommended remediation measures for the waste rock dumps at the Ronneburg mining site, which were derived from the integrated assessment of risks, costs, and benefits of different remediation options, are in compliance with the relevant guidelines regarding radiation protection and surface water/groundwater protection.

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Paper 5

SIMULATION OF LIBERATION AND DISPERSION OF RADON FROM URANIUM MILL TAILINGS

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Abstract

Radon emissions from uranium mill tailings may constitute a major source of environmental contamination and consequently a potential health hazard to the nearby population. Radon-222 gas is generated from the radioactive decay of ^{226}Ra present in the particles. When it is formed, radon is free to diffuse along the pores of the residues to the surface and escape to the atmosphere. A major concern in waste management and long term stabilization is to reduce radon emissions to near background levels. The common theoretical approach is to calculate the cover thickness that results in a radon flux lower than a stipulated and accepted value. The conceptual model is based on the principle of diffusion across a porous medium, which allows the mathematical description of the radon transport through the waste and the cover. The basic diffusion equations are used for estimating the theoretical values of the radon flux formed from the decay of the ^{226}Ra content in the waste material. The algorithm incorporates the radon attenuation with an arbitrary cover system placed over the radioactive waste. Alternatively, the thickness can be estimated for a cover that allows a lower radon flux. Once the radon is released into the atmosphere it is available for atmospheric transport by the wind. Radon atmospheric dispersion is modelled using a modified Gaussian plume equation that estimates the average dispersion of radon released from a point source representative of one or more uniform area sources. The model considers the median release point between all the areas contaminated. The concentration available for the dispersion is calculated from the total flux released. The dispersion can be simulated for different wind directions, with different wind velocities, as well as in the dominant wind direction.

1. INTRODUCTION

For the great majority of uranium mining activities uranium mill tailings represent the greatest potential source of environmental contamination. In the milling process the uranium ore is ground, promoting liberation and thus increasing the possibility of radon escaping to the environment. The milling process generates large volumes of tailings that are generally disposed of in ponds. The radionuclides present in the tailings, ^{226}Ra , ^{230}Th and ^{222}Rn , are a major concern for human health and the environment. They are not dissolved during the leaching process, which disturbs the equilibrium chains of the ^{238}U and ^{235}U decay families.

The principal radon isotope, ^{222}Rn , formed from the radioactive decay of ^{226}Ra , has a half-life of 3.82 d, which allows for a considerable period of time for migration before it decays into another nuclide. Radon generation may continue for thousands of years because of the long half-lives of ^{226}Ra and ^{230}Th present in the uranium mill tailings. Radon is an inert gas which emanates from the solid tailings particles and is free to diffuse to the surface of the pile and then escape to the atmosphere. Coming up from the ground it may become locally hazardous or it may be transported by the wind into the surrounding area, spreading the possible damage.

This paper proposes a two dimensional model for calculating the ^{222}Rn flux diffusion from radioactive waste disposal, giving the ^{222}Rn concentrations at a defined mixing height which will be the starting point of the atmospheric dispersion, as well as the dispersion in the prevalent wind direction.

2. RESULTS AND METHODS

2.1. ^{222}Rn diffusion

The basic diffusion equations may be used for estimating the theoretical values of the radon flux from the ^{226}Ra content in the tailings. Radon migration to the surface is a complex process controlled mainly by porosity (ϵ) and moisture content (θ), controlling the efficacy of the cover in attenuating the radon flux. The efficacy depends on the capacity of the cover material for keeping the diffusion so slow that radon decays to another non-gaseous nuclide, becoming trapped by the cover system.

The movement of radon in soil is characterized by the diffusion coefficient, D , which can be measured, either in a laboratory or in the field, or estimated by empirical correlations. The latter have the advantage of being

simple and easy to use with a minimal amount of information needed. A correlation using the fraction of saturation, m , is recommended:

$$D(cm^2/s^{-1}) = 0.07 e^{[-4(m-m\varepsilon^2+m^5)]} \quad (1)$$

Values for radon diffusivity in porous media may vary over a wide range of several orders of magnitude, depending on the porous material and particularly on its degree of water saturation. The generic diffusion equation can be represented by:

$$D \frac{\partial^2 C}{\partial x^2} - \lambda C + \frac{R\rho\lambda E}{\varepsilon} = 0 \quad (2)$$

The diffusion process occurs in a multiphase system, where the porosity is either filled with air or with water. If we apply the generic diffusion equation to each one of the phases (a air, w water filled pore space):

$$D_a \frac{\partial^2 C_a}{\partial x^2} - \lambda C_a + \frac{R\rho\lambda E_a}{\varepsilon - \theta} + \frac{T_{wa}}{\varepsilon - \theta} = 0 \quad (3)$$

$$D_w \frac{\partial^2 C_w}{\partial x^2} - \lambda C_w + \frac{R\rho\lambda E_w}{\theta} - \frac{T_{wa}}{\theta} = 0 \quad (4)$$

In these equations, D (m^2/s^{-1}) represents the radon diffusivity, λ the radon decay constant (s^{-1}), C (Bq/m^{-3}) the radon concentration in the pore space, R (Bq/kg^{-1}) the radium concentration in the material, ρ (kg/m^{-3}) the bulk density of the dry material, E the (dimensionless) radon emanation power coefficient for the pore spaces, ε the (dimensionless) total porosity, θ the (dimensionless) moisture content, and T_{wa} ($Bq/m^{-3}/s^{-1}$) the radon transfer rate from water to the air.

The solution of the diffusion equation for a homogeneous medium represents the flux release from the tailings to the surface, J ($Bq/m^{-2}/s^{-1}$). For a system without a cover we obtain [1]:

$$J_t = R\rho E \sqrt{\lambda D_t} \tanh\left(\sqrt{\frac{\lambda}{D_t}} x_t\right) \quad (5)$$

In this equation, x_t represents the tailings thickness. If we consider a two media problem represented by the tailings (t) and a homogeneous cover material (c), the solution of the diffusion equation can be represented by [1]:

$$J_c(x_c) = \frac{2J_t e^{-b_c x_c}}{\left[1 + \sqrt{a_t/a_c} \tanh(b_t x_t)\right] + \left[1 - \sqrt{a_t/a_c} \tanh(b_t x_t)\right] e^{-2b_c x_c}} \quad (6)$$

In this solution, $b_i = \sqrt{\lambda/D_i}$ (m) ($i = c$ or t), and $a_i = \varepsilon_i^2 D_i [1 - 0.74 m_i]^2$ ($\text{m}^2 \text{s}^{-1}$), where m is the degree of saturation of the soil.

From the preceding equations it is possible to obtain generic solution for the radon released in three main situations: flux directly diverted to the atmosphere without cover trapping, radon flux through a homogeneous cover and radon flux through a multilayer cover system.

Another possible approach is to calculate the thickness of the cover which allows a value stipulated and accepted for the radon flux. This can be done directly using the waste and cover parameters, mainly the ^{222}Rn coefficient diffusion, porosity and moisture, the ^{226}Ra content in the waste material and ^{222}Rn emanation, rearranging the last equation [1]:

$$x_c = \sqrt{\frac{D_c}{\lambda}} \ln \left[\frac{2J_t/J_c}{\left(1 + \sqrt{a_t/a_c} \tanh(b_t x_t)\right) + \left(1 - \sqrt{a_t/a_c} \tanh(b_t x_t)\right) (J_c/J_t)^2} \right] \quad (7)$$

2.2. Atmospheric ^{222}Rn dispersion

The model uses a Gaussian model of plume dispersion to account for the transportation of ^{222}Rn from the source area to a downwind receptor, and is represented by Pasquill's equation, as modified by Gifford [2]:

$$X = \frac{Q}{2\pi \mu \sigma_y \sigma_z} e^{\left[-1/2 \left(\frac{y}{\sigma_y} \right)^2 \right]} \left\{ e^{\left[-\frac{1}{2} \left(\frac{z-H}{\sigma_z} \right)^2 \right]} + e^{\left[-\frac{1}{2} \left(\frac{z+H}{\sigma_z} \right)^2 \right]} \right\} \quad (8)$$

This equation represents a Gaussian distribution, where X (Bq/m^3) is the radionuclide concentration, Q (Bq/s^{-1}) the strength of the source, and H (m)

the corrected source release height. Dispersion parameters, σ_y (m) and σ_z (m), are the standard deviations of the plume concentration in the horizontal and vertical directions, respectively. The standard deviations can be evaluated by the Pasquill–Gifford coefficients for flat rural areas or by the Briggs method for urban areas. Atmospheric transport is calculated at wind speed (height independent), u (m/s^{-1}), to a sampling position located at surface elevation, z , and transverse horizontal distance, γ , from the plume centre.

The concentration available for the dispersion is calculated from the radon flux. The contamination source is defined as an emission area or the sum of several areas, where the radon is diluted directly into the ambient air above the contaminated source zone. The wind speed for ^{222}Rn dilution should be matched to the average annual value through the mixing zone.

The contaminated area can be compared to an environmental compartment in which the ^{222}Rn emission is uniform over the whole area, characterized by its length and width. These values and the mixing height define the volume in which the concentrations are spatially homogeneous and instantaneously mixed.

The modified Gaussian plume equation estimates the average dispersion of radon released from a point source representative of one or several uniform area sources. This point represents the median release point between all the areas contaminated, with the same mass flux as all of the affected zones, and it should be located in the weighted centre of the total contaminated area.

The ^{222}Rn concentration dispersion is calculated from the release point, at the mixing height, for defined distances in each wind direction. The point source is located in the centre of a conceptual regular polygon area defined as the contaminated site. The concentrations along each direction are evaluated taking into account the respective mean wind velocity and the frequency of the occurrence. This will lead to different ^{222}Rn concentrations in the sectors defined by each direction in the collateral wind rose.

Considering the dominant wind direction exclusively, one assumes that all of the ^{222}Rn concentration will be distributed in that direction, being non-existent in all other directions. Another assumption is that the ^{222}Rn concentration is uniform over the volume defined by the central point source and the boundaries of the release area, and by the mixing height. Dispersion occurs outside this area.

2.3. A case study

The model's capacity was tested by simulating simple situations and checking its capacity to respond to variations in some of the parameters

involved. The model was applied to a specific contaminated site, the Urgeirica uranium mill tailings.

The necessary parameters were based on measurements made by ITN (Nuclear and Technological Institute) in the Urgeirica tailings piles [3]. Local meteorological data, namely wind velocity and frequency, were used for simulating the dispersion. The unknown parameters were estimated from available data.

The calculation proceeded in the following steps:

- (1) Estimation of the ^{222}Rn diffusivity in the tailings and in the cover;
- (2) Estimation of the ^{222}Rn flux to the air in the breathing zone;
- (3) Calculation of the ^{222}Rn concentration dispersion in the atmosphere in each wind direction, accounting for its intensity and frequency of blowing.

The contaminated site is composed of four different tailings disposals with a total area of approximately $110\,321\text{ m}^2$. It was assumed that no covering system exists over this area. The median point for the global area was defined by the arithmetic average of the medium point for each area and has the following coordinates: $x = 20\,612$, $y = 93\,326$.

The ^{222}Rn flux was calculated for the global area, conceptually defined as a regular octagon. Each sector has a characteristic average wind speed. The area contaminated is the same as the polygon area, and from it one can estimate the length of the polygon sides and the radius of the circumference. The air breathing or mixing height was defined as 1.7 m. The ^{222}Rn concentrations were calculated in each sector for this height. The total average flux estimated is $0.2042\text{ Bq/m}^{-2}\text{/s}^{-1}$.

The wind speed data in each direction (N–S–E–W–NW–NE–SE–SW) were obtained from the meteorological station at Nelas. The values fit into Pasquill stability class D (neutral), so they were chosen to estimate the typical dispersion coefficients. The dominant wind direction is NE.

The concentration at the breathing height in the dominant wind direction refers only to the polygon side that limits the respective sector.

The dispersed concentrations are different in each sector. We defined a 2 km distance from the centre for simulating the ^{222}Rn dispersion in each wind direction; the same distance was considered for the dominant wind direction (NE). The calculated dispersion results are shown in Figs 1, 2.

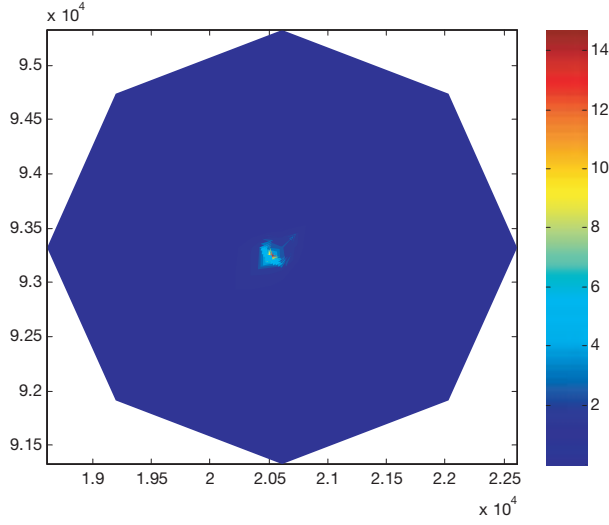


FIG. 1. Radon dispersion in each wind direction (Bq/m^{-3}).

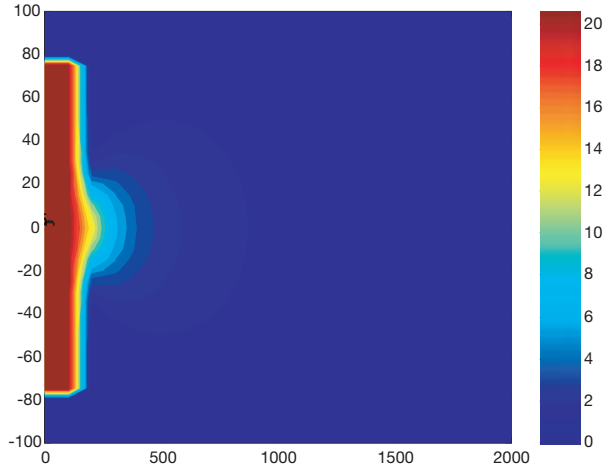


FIG. 2. Radon dispersion in the dominant wind direction (Bq/m^{-3}).

3. CONCLUSIONS

The work presented in this paper may be considered a first approach to modelling the dispersion of ^{222}Rn , thus allowing the assessment of exposure from uranium tailings piles. The obvious limitations are mostly related to the reduction of the source to a point. In addition, variations due to complex terrain topography cannot be modelled, as the model implicitly assumes a flat terrain. The wind velocity is constant with height and the dispersion is only two dimensional, although this direction rotates in the horizontal plane.

The difficulties that may be encountered when trying to obtain the data needed to characterize the contaminated material and also the variability of these data, both in space and time, should be noted. It should also be noted that the radon flux depends on the diffusion coefficient, which greatly varies with the material moisture and porosity. These parameters will vary over the year due to climate changes. It also depends on the radium content of the tailings, which may be different in each pile, and on the water retention, which is also a seasonal parameter with a wide range of values at the Urgeiriça site [3].

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Paper 6

RADON SURVEY IN OLD URANIUM MINES IN THE REGION OF SABUGAL, PORTUGAL

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Abstract

The Portuguese uranium industry — with a significant concentration of mines in the Guarda district — experienced a significant development period after World War Two, supported by military needs. Today all the uranium mines have been closed but radioactive releases may be a significant cause of environmental and radiological problems in and around the old uranium mining facilities. The main purpose of the present work is to assess outdoor and indoor radon levels, as well as radon exhalation rates, in the vicinity of six abandoned uranium mines (covering an area of about 70 km²) in the Sabugal region. The results show that the outdoor radon concentrations in the region range from 84 to 412 Bq/m³, while half of the indoor radon measurements were found to be above the EU recommended action level for existing buildings (400 Bq/m³). The exhalation rates from soils and tailings ranged from 6 to 323 mBq/m²/s⁻¹ during the first field campaign (July 2002), and from 7 to 1198 mBq/m²/s⁻¹ during the second campaign (August 2002). The highest values were confined to the mining areas or to their immediate proximity.

1. INTRODUCTION

Releases of radioactivity associated with uranium mining and milling operations are considered to potentially be a significant cause of environmental and radiological problems in and around mining facilities [1]. In Portugal, the physical and chemical processes that were used to extract uranium from ore, such as crushing and acid leaching, have produced considerable amounts of mill tailings. These tailings piles could concentrate high activities of ²²⁶Ra, reaching 50 kBq/kg in some spots [2], which is significantly higher than the average ²²⁶Ra activity value of 40 Bq/kg reported by UNSCEAR for natural soils [3].

The radioactive decay of ²²⁶Ra produces an inert radioactive gas, ²²²Rn, which has a half-life of 3.8 d. Despite its short half-life, a large number of ²²²Rn atoms produced in uranium mill tailings could reach the atmosphere, where

they decay through several short lived alpha emitters (like ^{218}Po and ^{214}Po) to the relatively long lived ^{210}Pb (half-life 22.3 a).

The radon exhalation flux to the atmosphere is strongly influenced by meteorological factors such as atmospheric pressure and rainfall rate, and by the characteristics of the tailings themselves, such as the ^{226}Ra concentration, moisture content and porosity.

The main purpose of the present work is the assessment of indoor and outdoor radon levels, as well as radon exhalation rates in and around six abandoned uranium mines (Rosmaneira, Coitos, Bica, Pedreiros, Carrasca, and Vale de Arca) in the Sabugal region.

2. SITE DESCRIPTION

The 70 km² which were surveyed are situated in the Sabugal region (Central Portugal). In this area the altitude ranges from 500 m to 800 m and the topography shows medium slopes. The watershed that separates the Tagus river basin from the Douro river basin crosses this area.

Geologically the region consists of Hercynian porphyroid monzonitic granites and muscovite-biotite granites, crossed by quartz veins as well as microdiorite and dolerite veins. The uranium bearing ore of Rosmaneira, Bica and Vale da Arca are of a vein type, with pitchblende, sulphides and secondary uranium minerals [4].

Between 1953 and 1959 five natural leaching installations were built at Urgeiriça, Valinhos, Rosmaneira, Bica and Vale da Arca, where 70 000 tonnes of low grade ore were treated with an efficiency of 55 to 75%, resulting in a production of approximately 47 tonnes of U_3O_8 [5]. The latter three mines are located in the studied area (Fig. 1).

During this survey, with the exception of the Bica mine where neutralization of acid groundwaters is still taking place, the other old mines are closed. All have tailings of varying amounts associated with them.

3. MATERIALS AND METHODS

3.1. Schedule

The radon survey was carried out during two field campaigns in July and August of 2002, respectively, covering an area of about 70 km² and comprising various types of measurements, including radon exhalation rates and outdoor and indoor radon measurements.

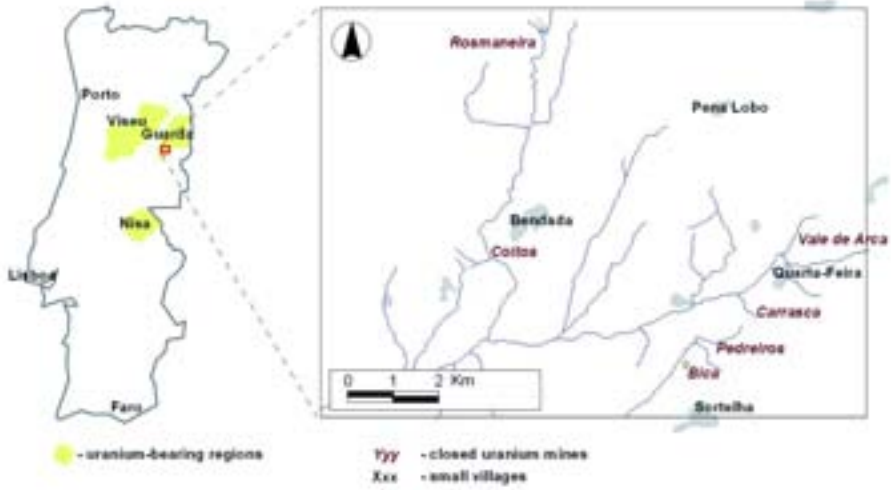


FIG. 1. Location of the six closed uranium mines considered in the region under study.

3.2. Exhalation measurements

For the radon exhalation measurements, open faced cylindrical containers (used as accumulation chambers) were placed on the soil surface with the open faces down. After an accumulation period that ranged between 17 and 25 hours, air samples were taken from the containers (small fans had previously been placed inside the containers and turned on immediately before the air was sampled to avoid possible radon stratification inside the chamber by the time of collection), filtered through a Millipore® type AA 0.8 mm filter and collected into a 125 cm³ scintillation cell (Lucas cell). Following a 3 hour equilibration period, ²²²Rn concentrations were measured using a photomultiplier counter (NOVELEC®).

The radon exhalation flux (Ex) was calculated using the following equation [6]:

$$Ex = \frac{C(t) \times V \times \lambda}{A \times (1 - e^{-\lambda \times t})} \quad (1)$$

where $C(t)$ is the radon concentration (Bq/m³), V is the volume of the container (m³), A is the surface area of the soil from which the exhalation takes place

(m^2), λ is the decay constant of radon (s^{-1}) and t is the accumulation time. The results are expressed in $\text{Bq/m}^{-2}/\text{s}^{-1}$.

During the first campaign 24 sites were monitored, covering a wide area in and around the tailings. For the second campaign, the survey network was reinforced with 11 more sites to better understand the boundary conditions of the tailings.

3.2.1. Integrated measurements

Integrated passive radon measurements were performed using solid state nuclear track detectors (LR-115 detector) placed inside protective boxes in the case of outdoor measurements. After an exposure period of about one month the films were etched and tracks counted on a spark counter.

A total of 44 outdoor detectors were distributed in pairs at 22 locations. In addition, 23 dosimeters were placed in local dwellings.

4. RESULTS AND DISCUSSION

4.1. Radon exhalation measurements

Overall it was found that radon exhalation rates ranged from 6 to $1198 \text{ mBq/m}^{-2}/\text{s}^{-1}$, the latter value being the highest obtained during the second field campaign (Fig. 2). Despite the observed differences between the two campaigns, the highest values were measured at the same sites: the Rosmaneira mine (307 and $1024 \text{ mBq/m}^{-2}/\text{s}^{-1}$ in July and August, respectively) and the Bica mine (323 and $1198 \text{ mBq/m}^{-2}/\text{s}^{-1}$ in July and August, respectively).

On further analysing the set of results it can be observed that the tailings areas exhibit, as expected, distinctly higher exhalation fluxes (with average values of 133 and $435 \text{ mBq/m}^{-2}/\text{s}^{-1}$ for the first and second campaign, respectively) compared to the fluxes obtained for the surrounding areas or soils (average values of 30 and $31 \text{ mBq/m}^{-2}/\text{s}^{-1}$ for the first and second campaign, respectively).

The repetitions performed during the second campaign allow us to identify situations like those illustrated in Fig. 3. Two measurement points at the Rosmaneira and Bica tailings reveal exhalation fluxes higher, by a factor of 19 and 17 respectively, than those measured at a short distance outside the tailings piles. A possible explanation for the higher values found during the second campaign could be the different weather conditions (Fig. 4).

During both field campaigns no rainfall occurred, but during the three days preceding the second campaign an 11 mm precipitation event was

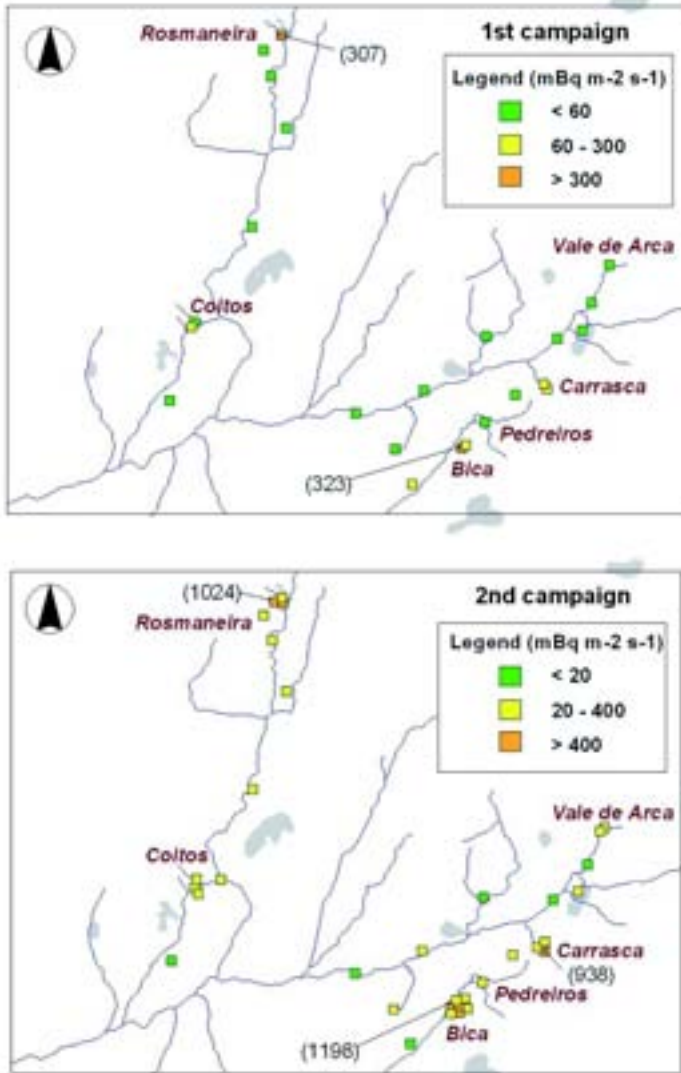


FIG. 2. Location of the exhalation measurement points in the region under study.

registered. This soil moisture increase could have augmented radon exhalation fluxes. According to Cothorn and Smith [7], up to a certain soil moisture level the positive effects of enhancement of the alpha recoil fraction can increase the exhalation rate by 50–250%.



FIG. 3. Two examples of identification of tailings boundaries.

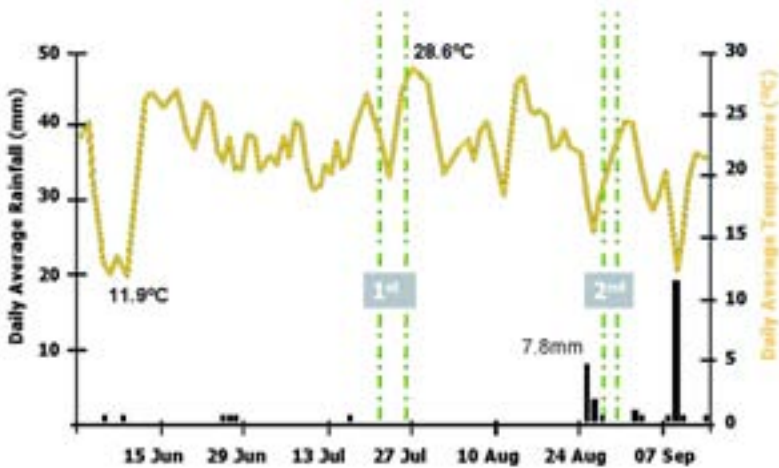


FIG. 4. Daily average rainfall and daily average temperature in the studied region. Data are from an udometric station located in Bendada (inside the studied area) and a meteorological station 15 km south-east of the area (Barragem da Meimoa). Both stations are included in the Water Institute (INAG) meteorological network.

It is interesting to note that the ^{222}Rn fluxes obtained from the Rosmaneira, Bica and Carrasca tailings piles are much lower than those reported by other authors [8, 9]. However, four measurements at three sites show values higher than the US recommended limit of $740 \text{ mBq/m}^{-2}/\text{s}^{-1}$ for uncovered tailings.

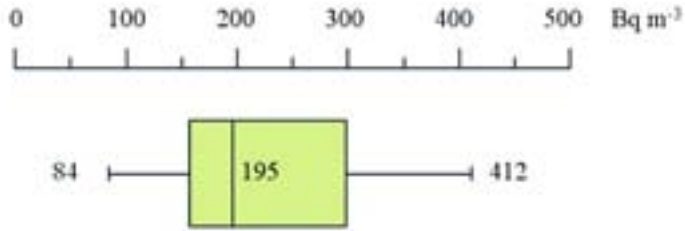


FIG. 5. Distribution of outdoor radon levels.

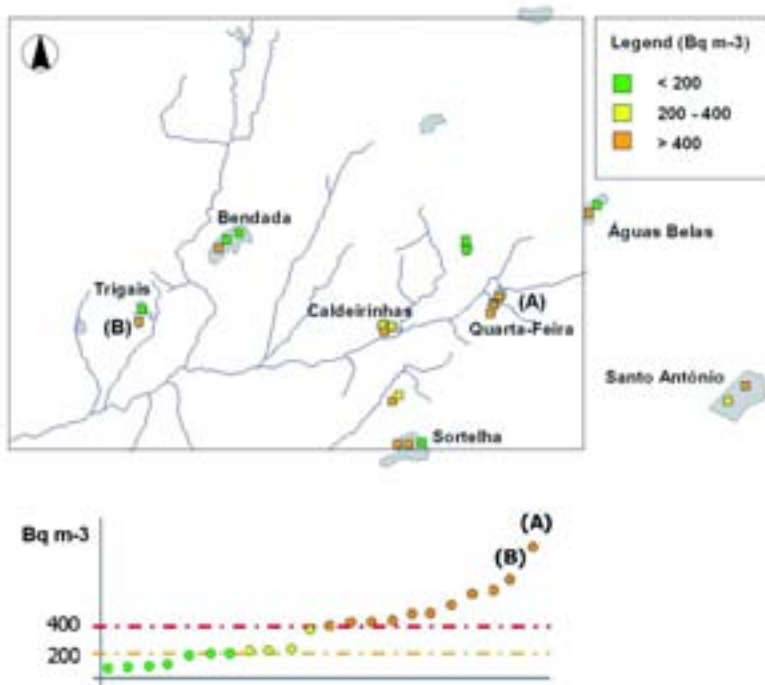


FIG. 6. Distribution of indoor radon concentrations.

4.2. Outdoor radon levels

Of the 44 dosimeters initially distributed, only 23 were recovered. However, since duplicates were used at each site only two locations produced no information.

The outdoor radon concentrations obtained ranged from 84 to 412 Bq/m³, with a median value of 195 Bq/m³ (Fig. 5). The highest values were found at six sites with no evident relation to any obvious sources (e.g. tailings).

4.3. Indoor radon levels

In half of the measurements of radon levels inside dwellings in small villages around the old mining areas (Fig. 6) values were found to be above the EU recommended action level of 400 Bq/m³ for existing buildings [10].

It seems that the geology under the dwellings plays an important role for indoor radon levels. As shown on Table 1, the highest values occur in locations that are crossed by important quartz veins, which in the region are usually associated with uranium mineralizations. This hypothesis should however be treated with care due to the small number of samples.

5. CONCLUSIONS

Concerning the exhalation rates it was found that the highest values are confined to the tailings piles areas, significant differences occur among the six tailings, and significant differences also occur inside each tailings area. The absence of local wind data, as well as the reduced number of recovered detectors, prevented a more consistent interpretation of the outdoor radon values.

TABLE 1. INDOOR RADON VALUES BY LOCATION AND GEOLOGY

Location	Geology	Number of measurements	Index
Quinta Clérigos	Granite	2	1
Bendada	Granite	3	1.7
Trigais	Granite	2	2
Águas Belas	Granite	2	2
Sortelha	Quartz vein	3	2.3
Caldeirinhas	Quartz vein	3	2.3
Bica Mine	Granite	2	2.5
Santo António	Quartz vein	2	2.5
Quarta-Feira	Quartz vein	3	3

Index = $\frac{\sum_{n}^{Class}}{n}$, where Class 1 = <200 Bq/m³; Class 2 = 200–400 Bq/m³; Class 3 = >400 Bq/m³; and n = number of measurements.

The 50% of indoor radon measurements above the EU recommended action level confirms this region (uranium bearing region) to be a radon-prone area where some kind of mitigation measures should be undertaken.

ACKNOWLEDGEMENTS

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Paper 7

WHAT COMES AFTER ACTIVE REMEDIATION: LONG TERM CHALLENGES AT URANIUM MINING AND MILLING SITES

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Abstract

Remediation plans for uranium mining and milling sites often tacitly assume that a certain amount of maintenance has to be carried out for almost unlimited time spans. The paper highlights the relevant issues and attempts to outline the conceptual, management and technical problems and challenges of maintaining institutional control over possibly hundreds or even thousands of years.

1. INTRODUCTION

Former uranium mining and milling sites often cannot be remediated to residual levels of contamination that are below concern and cannot be released for unrestricted use. Hence they have to remain under some form of institutional control. Residual contamination, mill tailings or low grade ore, and other hazards may remain for several reasons after cleanup is complete: technical limitations, economic feasibility, worker health and safety issues, or prevention of collateral environmental impacts. In particular, underground mine workings often cannot be cleaned up because access is impossible or too dangerous [1]. A compromise between social and economic cost and level of protection has to be found. With long lived radionuclides present, maintenance of institutional control is likely to be required for nearly unlimited periods of time. It may be noted that this does not seem to be such an issue in the case of hazardous waste landfills, which may contain compounds such as heavy metals that will exist forever.

The tasks and activities connected with maintaining institutional control over prolonged periods of time are often subsumed under the term 'stewardship' [2].

2. THE TIME HORIZON

A ‘steward’ is a person entrusted with the management of another’s property. In this sense stewardship means taking care of legacy sites. More specifically, the term has been applied to those instances or phases of a site where active remediation has been completed but some residual contamination remains, not allowing the free release of the site or land. The challenges faced by a ‘steward’ are illustrated in Fig. 1.

There are a variety of views of what may be considered ‘long term’, but a distinction can be made between stewardship strategies for the ‘societal’ planning horizon, i.e. for one generation (approximately 30 years), ‘archaeological’ (i.e. more than 1000 years) and ‘geological’ (i.e. more than 10 000 years) time horizons. Essentially stewardship is one phase of the life cycle of a site or facility.

Typically, approaches to long term stewardship share the following common elements: risk assessment, risk management and risk communication. Stewardship encompasses activities directed at a particular site but also has to maintain and develop the scientific understanding and technical know-how supporting the stewardship activities.

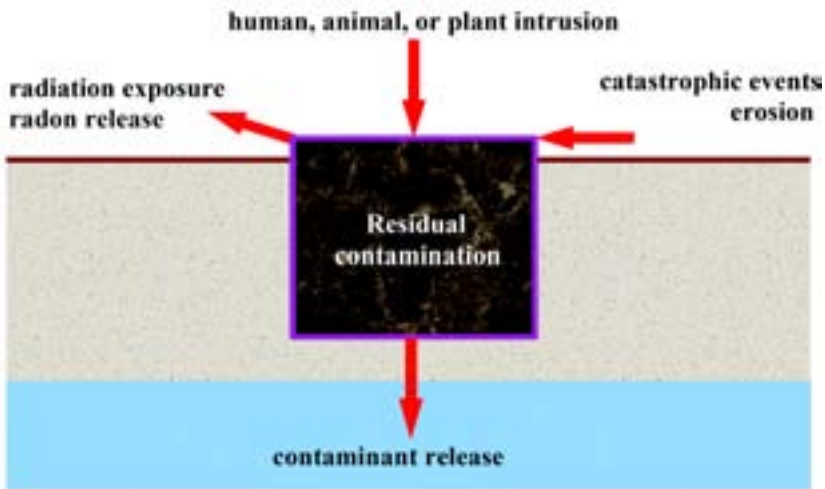


FIG. 1. Stewardship challenges.

3. LIFE CYCLE MANAGEMENT

3.1. An ethical question

Long term stewardship is one outcome of the struggle for sustainable development, which requires that our current activities do not impair the ability of future generations to live in a way they choose. It is interesting to note that we are the first generation to entertain such ethical notions and that we also hold ourselves responsible for the detriments caused by past generations. Future generations will enjoy the accumulated benefits from all previous generations and it can be argued that each generation should also carry some of the burden incurred by its predecessors. Hence, we could ask ourselves whether we really need to find ‘permanent’ solutions or whether we should not be able to leave some legacy to future generations that are likely to command as much knowledge and capability as ours and should be quite able to look after themselves.

3.2. Change in paradigms

In recent years a slow change in paradigms has occurred: a move away from treating environmental problems after they have occurred without positive feedback into the activities that have caused the problem, towards a more integrated management of human activities. This life cycle management approach aims to treat each stage in the life of a facility or site not as an isolated event but as one phase in its overall life (see Fig. 2). Thus planning does not only cover each stage but is a continuing activity, taking into account actual and projected developments and feedback mechanisms between different stages in the life cycle. Thus the remediation decision would be one step of the life cycle in a comprehensive planning process of, e.g. the fuel cycle, that provides the greatest benefit to cost ratio over the life of the facility in a holistic sense. However, long term stewardship must accept the many decisions that have been made previously in the life cycle and ensure that impacts on human health and the environment are minimized.

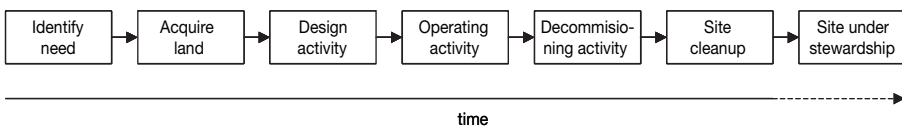


FIG. 2. Life cycle management.

The integration of planning for stewardship during the operational and remediation phases is not limited to physical actions. Other considerations may include the building up of trust funds for long term stewardship, avoiding the foreclosure of future options and taking contingencies into account when making decisions.

4. OBJECTIVES AND OUTCOMES OF REMEDIAL ACTIONS

The objectives and outcomes of remedial actions have a direct and lasting effect on the level of long term stewardship required at a site. Reference [3] stipulates that “remediation measures shall be justified by means of a decision aiding process requiring a positive balance of all relevant attributes relating to the contamination. In addition to the avertable annual doses, both individual and collective, other relevant attributes shall be assessed”.

Decision makers are faced with three fundamental choices for the intended remedial action [4]. They must decide whether they will:

- (1) Leave the site undisturbed while establishing a monitoring scheme for determining the evolution of the site. This option relies on natural processes to prevent significant exposure. The entire process needs to be carefully monitored so that alternative action can be initiated if required.
- (2) Contain or restrict the mobility of the radioactive contaminants. Such technologies aim to immobilize the contaminants inside the area where they already exist, reducing the potential for further migration or entry into active exposure pathways.
- (3) Remove the radioactivity and other contaminants from the site using an appropriate treatment method. Such treatment technologies aim to extract, concentrate and then safely dispose of the contaminants at another location.

The need for remediation and judgement as to acceptable residual contamination levels are usually driven by society's perception of the balance between the costs of measures and the benefits obtained. As has been discussed elsewhere [5, 6], there is a certain ‘window’ for decision making, bounded by minimum required benefits and maximum allowable expenditure. Expenditures typically increase exponentially with the lowering of residual risks.

Remediation typically proceeds in an iterative fashion and end states emerge as the de facto result of multiple interim actions. Measurement of the success of remediation is still a developing science.

While, obviously, contaminant removal is a permanent solution for the site in question, any engineered solution to contain residues at some other place or to reduce exposures will only have a limited useful lifetime. Natural forces will gradually degrade structures such as barriers or covers. Stewardship and life cycle management will have to take this into account. This uncertainty over the long term effectiveness of remediation solutions requires provisions for monitoring [6], periodic performance assessment and, if required, maintenance, i.e. the establishment of a stewardship programme. It is this uncertainty that mainly requires long term stewardship. While making remediation decisions it is important to consider long term stewardship issues and obligations explicitly when examining remedial alternatives and implementing a final remedy [7].

Measurement of the success of remediation is still a developing science. Many times the determination of when remediation is complete is based on when the regulator certifies, or in some way designates, that the remedial actions taken have met the established remedial objectives. The determination of when remediation is complete and long term stewardship begins may differ between Member States and may well vary for different types of sites within a Member State.

5. TECHNOLOGICAL CHALLENGES

Long term stewardship requirements pose a range of technological challenges. The ultimate goal, however, is the installation of engineering solutions that obviate stewardship. There is a high degree of interaction between remediation technology and strategy deployed and the resulting stewardship needs. It will likely have to be accepted that finding really ‘permanent’ solutions will be impossible. Learning from natural processes such as diagenesis and erosion behaviour could be a way forward that could be summarized in the paradigm engineering with nature, not against it. Compatibility with the surrounding geology and geomorphology, thus achieving low relief energy (shallow slopes), and compatibility with surrounding ecosystems, including the hydrologic regime and vegetation system, may be an answer. Natural ‘analogues’ have long been used to overcome the limitations inherent to short term laboratory or field experiments for assessing the long term behaviour of human-made structures.

From a geomechanical point of view, shallow slopes contribute to achieving low relief energy (Fig. 3). This is what natural geological processes achieve over millennia and our engineered structures may have to learn from

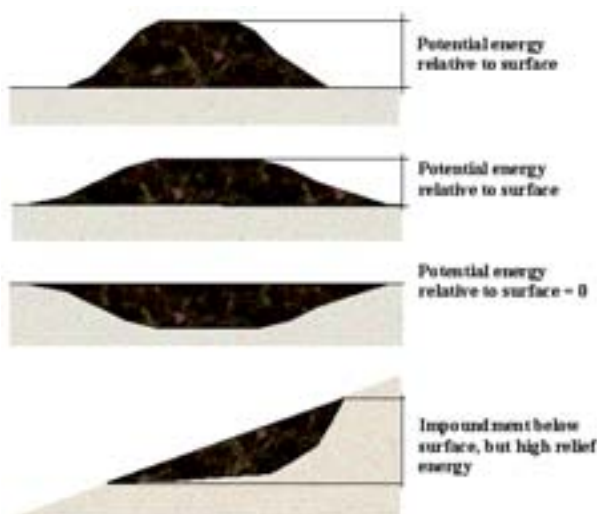


FIG. 3. Illustrations of the concept of inherent potential energy in the design of impoundments.

observation of the evolving geomorphology and slopes in the environment surrounding a site.

Natural evolution of soils and diagenesis also give insights valuable to the development of long term management plans. The contaminated material will not remain unchanged in the long term and assessment of its evolution will support confidence in the project if diagenesis improves retention of contaminants.

The ecosystem around a remediated site is the result of a series of processes that have been ongoing for centuries or millennia and are being shaped by a wide variety of initial conditions and contributing factors, such as the initial rock type, climatic evolution, surrounding flora and fauna, etc. The result is a (dynamic) equilibrium between soil type, vegetation cover and climatic conditions. Any attempts to reconstitute an ecosystem at the site, such as revegetation, need to be as compatible as possible with the surrounding ecosystem(s). The final use of the site needs to be compatible with the ecosystem in order to minimize pressure on the site due to human use.

6. LONG TERM STEWARDSHIP STRATEGIES

Public interest may call for low stewardship needs, but there may be economic constraints to achieving this. In many Member States current treasury rules and political domination of budgeting make it difficult to provide for the necessary long term funding security of stewardship programmes. Alternative instruments, such as trusts and bonds, have been proposed.

Stakeholder participation in the development of stewardship plans is considered a key factor in their success, as it has the potential to create ownership in the chosen solution. The definition and delineation of the 'public' and the 'stakeholder' are neither straightforward nor unequivocally accepted [5, 8]. Indeed, any one individual can be both, a general member of the public and a stakeholder, depending on whether the private, political or professional aspect of the life is concerned.

Maintaining institutional control is the major issue and land use planning and associated control instruments will be a major instrument. A variety of control instruments are being discussed, such as zoning regulations, deeds and easements, but all assume that the system of governance will not change dramatically and that somebody will be there to enforce them. While there are examples of forms of governance surviving for several centuries, recent history has shown dramatic changes even in traditionally more stable regions of the World. Hence, alternative strategies, such as 'taboos', 'stigmas', or sustained uses, e.g. cemeteries, are being discussed.

7. RECORD KEEPING AND KNOWLEDGE MANAGEMENT

Effective stewardship requires specific skills and knowledge. Retaining these has proven difficult as mining and milling sites undergo the transition from operation to remediation to stewardship. The diminishing number of trained people in the nuclear industry is a recognized problem. Also surprisingly, the maturing market for environmental services has led to a decrease in the availability of skilled personnel.

There is a general notion that future generations will command more knowledge and capability than the present generation. However, as is evident from many archaeological 'mysteries', such as the true purpose and design objectives of some archeological sites, and 'lost' production technologies such as the composition of certain medieval glass stains, some critical knowledge and insight might and will get lost.

Records are an essential basis for a successful stewardship programme, but there is no consensus as to which records are really important and how they

should and can be preserved. Different stakeholders will have different information needs that will also change over time.

While archiving and maintenance procedures for paper records are fairly well established and time proven, electronic records are a major concern. Many systems that were once considered high technology simply no longer exist and records stored in the respective format become lost. Also, most electronic records are physically much more vulnerable than paper records. No final solution to this problem has yet emerged.

As the example of the Egyptian hieroglyphics illustrates, preservation of the physical record is only one element in record keeping. Being able to read and interpret the record correctly is the essential complementing element. Societal and cultural context may be needed to understand the contents and purpose of records. As we cannot know what kind of society will require our records, it is virtually impossible to cater to its needs.

8. CONCLUSIONS

The technological and organizational challenges posed by long term stewardship programmes are reasonably well understood, but there are no satisfactory and ‘proven’ solutions to many of these problems. The present generation may have to be content to provide state of the art technical solutions while accepting that future generations may have different judgements. Our engineered solutions may well become the future contaminated sites.

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Paper 8

REMEDIATION OF THE LOW LEVEL RADIOACTIVE WASTE TAILINGS POND IN KOWARY (POLAND)

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Abstract

The paper presents in a condensed form the characteristics of the tailings pond in Kowary (Poland), the plan for remediation, as well as the most essential elements of the remediation technology applied. Post-remediation investigations indicated that the remediation results are consistent with planned indicators. The pond was eliminated from the surface of the land, dams were stabilized, the radiation was reduced to background level, i.e. <300 nSv/h, the surface and groundwaters from the area of the settling tank were drained and taken outside for later treatment in the existing water treatment system. The entire area was cleaned up and — due to vegetation development — blended into the landscape.

1. INTRODUCTION

An intensive search for uranium ore in Poland was initiated right after World War Two. The search and the initial mining works have left various contaminated sites and waste dumps. There were only five mines where the uranium ore was sufficiently concentrated to be of practical use. Over time, despite the low grade of the uranium deposits, their exploitation was continued and a processing line for chemical treatment of the ore and a tailings pond for the waste from the process were built in Kowary. For six years, the ore was brought there from the other mines to be processed into concentrate [1, 2]. Kowary was the only tailings pond of its kind in Poland.

2. LOCATION AND HISTORY OF THE KOWARY TAILINGS POND

The Kowary tailings pond is located in the Sudet Mountains in the southwestern part of Poland. It was constructed in 1967 to receive the waste from uranium concentrate production. It was placed close to the existing uranium ore mine in the town of Kowary.

The history of the mine reaches back to the Middle Ages. Initially it was an iron ore and silver mine. Uranium ore was discovered by the Germans prior to World War Two, and that is when trial exploitation was initiated. After World War Two the mine fell under Soviet jurisdiction. At first raw unprocessed ore was shipped back to the USSR. With time, however, when richer deposits of uranium ore were discovered, a dedicated mill became a viable proposition, rather than shipping the ore to the USSR. Therefore a processing line for low grade ore treatment and production of uranium concentrate was built. The production waste was pumped to the tailings pond. In 1972 the mine and uranium concentrate production were halted. However, the partially filled tailings pond was left behind. The remaining facilities were adapted for other experimental processes and the liquid waste from those were also disposed of in the tailings pond. In 2000 the tailings pond was shut down and the mining site, along with the tailings pond, was handed over to Wrocław Technical University. Due to natural forces, the condition of the tailings pond deteriorated [3, 4]. Consequently, Wrocław Technical University initiated permanent remediation. Using its own funds and research resources, the university conducted basic research and drew up a remediation concept. Government funds were obtained to carry out thorough research and to design the remediation project. This work became the basis for obtaining funding from the EU under the PHARE mechanism, which was essential for completion of the project [5–7]. The contractor chosen was the German company GEOS.

3. TECHNICAL DATA ON THE KOWARY TAILINGS POND

Following are the main characteristics of the Kowary tailings pond [3]:

- (a) Location: on the surface of a mountain slope with 10–20° incline, mean 12°;
- (b) Base: stony–clayey weathering layer of granite and gneiss;
- (c) Surface area: 2.3 ha (including the dams);
- (d) Dam: stony–clayey material, clay sealing in places, maximum height 12 m, river flowing at the foot of the highest part;
- (e) Impounded material: chemically milled treated rocks, mean grain size 0.25 mm;
- (f) Moisture content between 14.2 and 54.1%, mean 24.5%;
- (g) Impounded mass: 250 000 t;
- (h) Free water volume over the deposit: 5000 m³.

4. POTENTIAL AND ACTUAL HAZARDS

A range of potential and actual environmental, geotechnical and radiation hazards are associated with the Kowary tailings pond and its dams:

- (a) Unreliable stability of the dam ($=0.95-3$);
- (b) Danger due to spill-out of tailings;
- (c) Semi-fluid consistency of the impounded material;
- (d) Chemical composition of the impounded material;
- (e) Direct radiation of 300–4000 nSv/h [2, 8];
- (f) General erosion under the given climatic conditions;
- (g) Acute erosion from the adjacent river;
- (h) Geotechnical instability of the open underground mine workings.

It should be noted that there are residential dwellings only 20–50 m away from the dam [4].

5. GOALS AND OBJECTIVES OF THE REMEDIATION PROJECT

The following goals and objectives were formulated for the remediation project [2, 3, 9]:

- (a) Lowering of the direct radiation to background levels (as measured next to the tailings pond);
- (b) Stabilization of the retaining dam;
- (c) Management of the water from atmospheric precipitation, still existing mine drainage, drainage water from the consolidation of the tailings and seepage from the pond;
- (d) Neutralization of the acid tailings at the surface;
- (e) Final encapsulation of the tailings pond;
- (f) Treatment of the tailings pond drainage waters;
- (g) Contouring and revegetation to blend the impoundment into the surrounding landscape.

Care was taken not to use any materials in the remediation process that could lead to further deterioration of the environment.

6. IMPLEMENTATION OF THE REMEDIATION PROJECT

6.1. Overview

The project was implemented in five stages: preparatory work, design work, the actual remedial action, acceptance testing, and preparation for monitoring [10–12]. A three person team of specialists, along with a ten person community committee, coordinated the work.

6.2. Preparatory work

The preparatory work included the following:

- (a) Researching the formal legal conditions (regulating the ownership, site planning, legal situation of the tailings pond, that is whether it falls under the jurisdiction of mining, construction or water laws, etc);
- (b) Assessing the load capacities of roads and bridges, distance involved, etc. with the view to transporting materials;
- (c) Determining the existence of any area development plans;
- (d) Assessing the general opinion of the local community concerning the planned undertaking;
- (e) Preparing topographical maps and plans;
- (f) Installing measurement and control instruments (benchmarks, piezo-meters, piezo-inclinometers).

6.3. Site surveys and laboratory tests

6.3.1. Scope

The objective of these tests was to provide information about the:

- (a) Properties of the tailings pond base;
- (b) Construction and condition of the dams;
- (c) Physical, chemical and microbiological characteristics of the tailings and overlying waters);
- (d) Hydrologic balance of the pond area;
- (e) Direction of water flow and contaminant migration;
- (f) Magnitude and distribution of direct radiation fields and the respective background;
- (g) Effect of the underground mine workings;
- (h) State of the environment prior to the remediation.

The investigations were carried out by six teams working together:

- (1) The radiological investigations team;
- (2) The chemical investigations team;
- (3) The geotechnical investigations team;
- (4) The geological and mining investigations team;
- (5) The hydrogeological investigations team;
- (6) The biological and environmental investigations team.

The team designing the environmental assessment worked independently of the others.

6.3.2. *Field surveys*

The field surveys consisted of:

- (a) Geological, hydrological and environmental mapping;
- (b) Geological drilling and sampling;
- (c) Determination of filtration coefficients in boreholes;
- (d) Measuring of radiation fields in boreholes and on the the surface (1 m height);
- (e) Measuring the concentration of radon in soil air and above the surface;
- (f) Measuring the flow rates of surface waters and drainage;
- (g) Dendrological assessments;
- (h) Geoelectric mapping;
- (i) Localization of underground workings by georadar.

6.3.3. *Laboratory tests*

The laboratory tests consisted of the determination of:

- (a) Soil mechanics parameters for the base, dam, and the impounded materials;
- (b) The petrographic properties;
- (c) The filtration coefficients;
- (d) The chemical composition of the impounded material and waters, with particular emphasis on radionuclides and heavy metals;
- (e) Microflora species.

6.4. Analysis of results

The following conceptual research models have been designed based on the measurements, site surveys and laboratory tests:

- (a) Radiological, i.e. emissions of radionuclides: direct radiation of the waste material, radon gas into the atmosphere, radioactive dust by wind erosion, radionuclides through seepage into ground and surface water;
- (b) Topographical;
- (c) Geotechnical;
- (d) Hydrogeological;
- (e) Geochemical.

The preparatory work and research lasted 2.5 years.

6.5. Remediation strategy

The remediation strategy stipulated the following tasks:

- (a) Construction of a waste water treatment plant;
- (b) Cutting off of all influxes to the tailings pond (surface waters, industrial waste waters);
- (c) Construction of a peripheral drainage system that cuts off the influxes of precipitation waters into the tailings pond;
- (d) Construction of a collecting well for the drainage of waters from the tailings pond, with the option of pumping the water to the treatment plant or into the river;
- (e) Pumping off of the free water from the tailings pond;
- (f) Uprooting of trees from the area of the so-called beach and the tailings pond's crest;
- (g) Construction of a multi-layer bottom liner and surface cover consisting of (Fig. 1):
 - Covering the bottom of the tailings pond with lime (thickness 0.6 cm);
 - Covering the bottom with a geotextile (type 300 g/m²);
 - Successive emplacement of a geogrid and covering it with dolomite aggregate ($d = 10\text{--}50$ mm), the thickness of the dolomite layer being 0.3 m;
 - Covering the dolomite layer with a geotextile (250 g/m²);
 - Material from the tailings beaches and the northern batter;
 - Bentomat;
 - Dolomite;

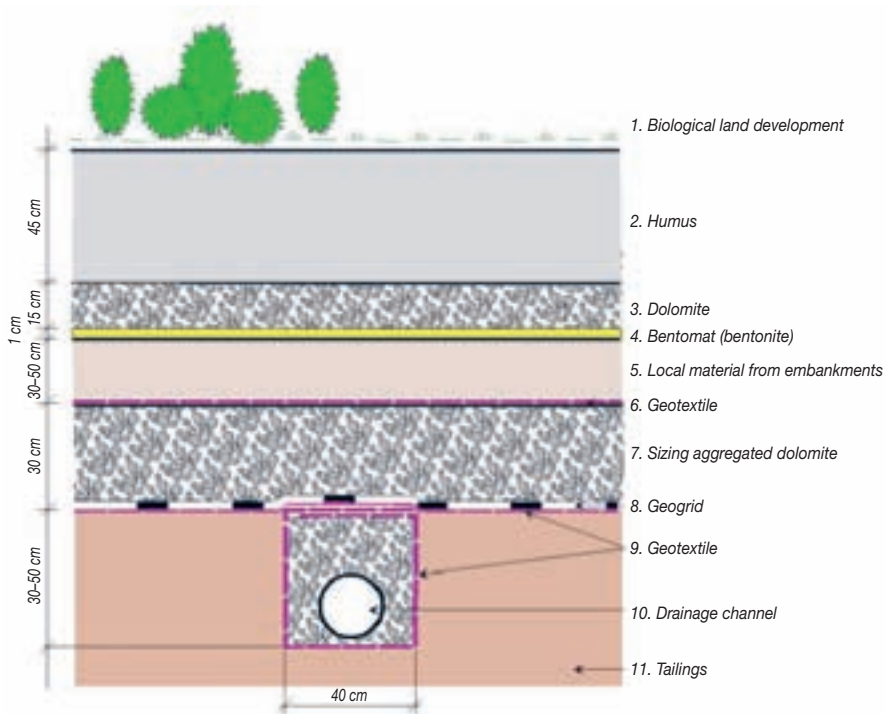


FIG. 1. Schematic surface cover design and horizontal internal drainage channel.

- Humus;
- Development of a vegetation cover;
- (h) Installation of an internal peripheral drainage made of drainage pipes on the bottom of the tailings pond, inside of the dam;
- (i) Installation of drainage pipes within the dolomite layer and directing them outside the tailings pond to the collecting well;
- (j) Removal of the material with increased radiation levels from the surface of the tailings pond and placing it in excavations inside the pond;
- (k) Construction of a surface drainage inside the tailings pond;
- (l) Placement of horizontal insulation made of a horizontal bentomat;
- (m) Levelling works to direct the slope of the backfilled pond toward the surface drainage (inside the pond);
- (n) Construction of a retaining wall made from gabions on the shore of the river and at the foot of the dam;
- (o) Improvement of the stability of some sections of the dam by decreasing the slope of the batter (embankments at the foot or shearing the batter);
- (p) Covering with a layer of topsoil;
- (q) Sowing grass and planting shrubs.

Dolomite ballast was chosen because it:

- (1) Screens the direct radiation;
- (2) Neutralizes and buffers the pH in the tailings pond porewaters;
- (3) Provides an adequately strong foundation in conjunction with the geogrids;
- (4) Provides horizontal drainage for seepage and consolidation waters.

No vertical drainage systems for dewatering the tailings were foreseen. The properties of the tailings and the impoundment are such that consolidation will take place without them. It was estimated that the settling of the tailings will not exceed 0.4 m, thus all drainage installations are expected to remain efficient.

The results of the remediation efforts are shown in Fig. 2.

7. SUMMARY AND CONCLUSIONS

The techniques were selected for their efficiency, their simplicity in implementation, as well as their low price.

After the required design had been established local mineral material was used in the project. Local materials were selected because they satisfy the quality and the geotechnical stability requirements, there is no need to purchase soil, implementation is quick, and there are no unfavourable microflora in the ground.

Bentomats were used as horizontal sealing layers as the nearest deposit of clay is 40 km away from the site, and thus shipping it would be too expensive. Moreover the bentomats are easier and quicker to deploy.



FIG. 2. The Kowary tailings pond before (left) and after (right) remediation.

The final tests indicated that the dam is stable, works properly, and the radiation was lowered down to background levels (below 300 nSv/h), with the exception of three places on the surface with an area of up to 10 m², where 600–800 nSv/h were measured. Warranty corrections were made there that consisted of spreading some additional fine grained dolomite ballast. The grass and the shrubs are growing very well.

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Paper 9

DECOMMISSIONING CANADIAN URANIUM MINES AND MILLS: REGULATORY FRAMEWORK, CLEANUP CRITERIA AND OBJECTIVES

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Abstract

The objectives of decommissioning uranium mines and mills are to remove, minimize and control potential contaminant sources, achieving an end state property that is stable and safe for humans and non-human biota over the long term. Achievement of these qualitative decommissioning objectives is defined in relation to the existing provincial and federal environmental quality objectives and, where objectives are not available, site specific benchmarks are derived. To illustrate the joint regulatory approach the paper describes the final decommissioning plans and objectives for a uranium mine and mill facility that operated for 22 years. The decommissioning activities include the reclamation of tailings, open pits, underground mines and waste rock piles, and the final disposition of all surface infrastructures, including the mill. Cleanup criteria and water quality objectives have been established for decommissioning the site, through regulatory review and public consultation, taking into account the anticipated future use of the site. Modelling and risk assessment have been performed to predict long term ecological effects and potential effects on human health. The results of all the analyses, and in particular the uncertainties in the modelling predictions, were then used to identify follow-up requirements for the project and to identify any contingency measures if the environmental monitoring indicates that the decommissioning objectives may not be achieved or that the potential environmental effects may exceed those predicted.

1. INTRODUCTION

The Canadian uranium mining and milling industry has evolved over time as our understanding and concern for impacts on both human and non-human biota have increased. At present the only active uranium mines and mills are located in the province of Saskatchewan, Canada.

The first uranium mines, established in the early 1940s and 1950s, were regulated to varying degrees and did not consider decommissioning or environmental impacts in the mine plans. As a result, several of these mines have had

little or no decommissioning completed despite their closure several decades ago. Several of these sites have legacy issues and will require costly remediation efforts and institutional controls over indefinite periods of time.

More recent mines, developed in the late 1970s to early 1980s, have been regulated by both provincial and federal regulatory bodies since project inception. They have been subject to detailed regulatory reviews and approvals, and more extensive public consultation throughout their project history. The owners of these mines, unlike the earlier mines, are required to maintain decommissioning plans and financial guarantees prior to and through the project life. These are subject to regulatory approval by both federal and provincial regulatory bodies.

The Cluff Lake uranium mine and mill was assessed and developed in the late 1970s and operated from 1980 to 2002. It is the first of the recently developed uranium mines in Canada to proceed with final decommissioning. Over the past three years the decommissioning proposal for the Cluff Lake mine has been the subject of a detailed environmental assessment, which has included the development of decommissioning objectives.

2. REGULATORY FRAMEWORK

2.1. Overview

The Nuclear Energy Act (NEA), the Nuclear Safety and Control Act (NSCA) and the Nuclear Liability Act (NLA) are the cornerstones of Canada's legislative and regulatory framework to ensure the safety of the nuclear industry in Canada.

The NSCA establishes the Canadian Nuclear Safety Commission (CNSC), which is comprised of the Commission (quasi-judicial tribunal that makes licensing decisions), and the CNSC staff. The CNSC staff prepares recommendations to the Commission, exercises delegated licensing and authorization powers, and assesses licensee compliance with the NSCA, associated regulations, and licence conditions. The CNSC's primary mission is to regulate the development, production and use of nuclear energy and materials, to protect the health, safety, security and environment, and to meet Canada's international commitments on the peaceful use of nuclear energy.

The CNSC's regulatory regime covers the entire nuclear substance life cycle, including uranium mining and milling in Canada.

In addition to regulatory oversight provided by the CNSC the uranium mines and mills are subject to federal regulatory requirements by 'Environment Canada' in matters relating to environmental assessments and

effects and effluent releases, and ‘Fisheries and Oceans’ in matters relating to fisheries and fish habitat. In the province of Saskatchewan (the location of all active uranium mines and mills in Canada), ‘Saskatchewan Labour’ provides regulatory oversight in matters of occupational mine safety while ‘Saskatchewan Environment’ regulates environmental matters, including pollutant control facilities, water treatment, forestry and aggregate management. Provincial requirements are generally imposed through the establishment of surface lease agreements with the mine operator. Since 2002 the CNSC and the two provincial Saskatchewan departments have been successfully harmonizing their respective regulatory roles regarding the Saskatchewan uranium mining industry.

2.2. Licensing process

Each of the project phases for a uranium mine or mill, including site preparation, construction, operation, decommissioning and abandonment, are subject to licensing review and approval by the CNSC. Licence applications are assessed for compliance with the NSCA and associated regulations. In addition, prior to proceeding with a licence review and approval, the proposed project is reviewed to determine whether and what type of environmental assessment (EA) is required, based on the requirements of the Canadian Environmental Assessment Act (CEAA) and its regulations. The decommissioning of uranium mines and mills will generally trigger a federal environmental assessment under the CEAA. While formal EA processes are in place provincially, their EA normally covers the complete life cycle and is not repeated once decommissioning is proposed. However, the province does participate as a reviewer in the federal EA process for decommissioning.

Under the CEAA, for decommissioning uranium mines and mills the CNSC is normally the responsible authority (RA) and is tasked with ensuring the completion of an environmental assessment and determining whether the project can be completed with no significant adverse environmental effects. It may delegate the completion of supporting technical documents to a third party, often the proponent, as they have the greatest knowledge of their proposed activity. However, these supporting documents are completed with the review and oversight of the RA. The federal and provincial authorities participate in the review process by providing expert advice where applicable, and the EA also includes public consultation processes by both the proponent and the regulators.

The RA is responsible for the determination of the environmental significance of the project and the completion of the final EA report. This report is submitted to CEAA for a formal public review process and final

determination. The project can only proceed to a formal CNSC licence application and assessment if it receives approval by CEAA.

2.3. Environmental assessment process

2.3.1. Overview

An EA provides a systematic approach to identifying the potential environmental effects of proposed projects. Where unacceptable adverse environmental effects are identified, decision makers can modify their plans to minimize or eliminate these predicted effects [1]. The final determination of the significance of the environmental effects is based on the project with mitigative controls in place.

The following provides the approach used in conducting an environmental assessment for the Cluff Lake decommissioning project, located in northern Saskatchewan.

2.3.2. Existing environment

The assessment process began with the identification of all contaminant sources on the project and the existing environmental conditions. Effects on environmental components — air quality, ambient radiological levels, hydrology, geology, terrestrial ecology, aquatic ecology, human health and land use were considered. This existing environment characterization was then used as the benchmark for determining the incremental effects associated with the decommissioning activities and the post-decommissioning recovery of the environment.

Underground and open pit mines, as well as a mill complex, operated at the Cluff Lake site. Issues surrounding surface contamination, acid rock drainage, leachate from tailings management areas, and downstream impacts from treated effluent releases all need to be addressed as part of decommissioning. The greatest predicted and observed environmental effect, outside of the mine's immediate footprint, is associated with Island Lake, the first lake located approximately 500 metres downstream of the effluent release point, where treated effluent was discharged over the site's 22 year operational period.

2.3.3. Valued ecosystem components

Valued ecosystem components (VECs) were identified and used as representative indicator ecosystems or biota for modelling the potential ecological implications of the proposed decommissioning activities. A VEC can be

defined as an environmental attribute or component perceived as important for social, cultural, economic or ecological reasons, and identified through consultation with affected people and through scientific opinion.

For the Cluff Lake project, the selected VECs (Table 1) consisted of biota important to northern Saskatchewan's local residents, as well as those considered to be ecologically significant or representative for assessing primary contaminant pathways.

2.3.4. Decommissioning objectives

2.3.4.1. General objectives

Decommissioning objectives were established to facilitate the evaluation of alternative means of carrying out the decommissioning project. The qualitative objectives of the decommissioning activities to be conducted at Cluff Lake are to ensure that:

- (a) The environment is safe for non-human biota and human use;
- (b) Long term adverse effects are minimized;
- (c) All constructed structures are removed or stabilized;
- (d) The reclaimed landscape is self-sustaining;
- (e) Restrictions on future land use are minimized.

Achievement of these qualitative decommissioning objectives was defined in relation to existing provincial and federal environmental quality and radiation protection objectives and where objectives were not available site specific benchmarks are derived. Generally, these objectives represent concentrations or limits below which significant adverse effects on human and non-human biota are unlikely. In addition, objectives in relation to long term institutional controls were also defined.

TABLE 1. SUMMARY OF THE VECs FOR THE CLUFF LAKE DECOMMISSIONING PROJECT

Environment	VEC
Aquatic	Pond weed, phytoplankton, benthic invertebrate, zooplankton, northern pike, lake whitefish, white sucker
Terrestrial	Herbivores (woodland caribou, moose, ptarmigan, snowshoe hare), omnivores (black bear, muskrat, ducks including scaup, mallard and merganser) and carnivores (wolf, bald eagle and otter)

In consultation with provincial and federal authorities and the public these objectives, and the appropriate locations and timeframes to achieve the objectives, are established through consideration of spatial–temporal relationships of the identified contaminant sources.

2.3.4.2. Water quality decommissioning objectives

Table 2 summarizes the water quality objectives established for the Cluff Lake decommissioning project. The Saskatchewan Surface Water Quality Objectives (SSWQOs) were adopted as decommissioning water quality objectives, with the exception of iron which is naturally elevated in the project area.

TABLE 2. SUMMARY OF SSWQOs

	Unit	SSWQO	Island Lake	Other lakes and streams	Flooded pits ^a
As	µg/L	50	50.11	50	50
Ba	µg/L	1	1	1	1
Cd	µg/L	1	1	1	1
Cr	µg/L	20	20	20	20
Cu	µg/L	10	10	10	10
Fe ^b	µg/L	1	1	1–7.3	7.3
Pb	µg/L	20	20	20	20
Hg	µg/L	0.1	0.1	0.1	0.1
Ni ^c	µg/L	(c)	(c)	(c)	(c)
Se	µg/L	10	10	10	10
Ag	µg/L	10	10	10	10
Zn	µg/L	50	50	50	50
²²⁶ Ra	Bq/L	0.11	0.11	0.11	0.11
U ^d	mg/L	—	(d)	(d)	(d)
Mo ^b	µg/L	—	500	73	500
Co ^e	µg/L	—	20	20	20

^a Flooded pits — Objectives apply to the upper 50% of the water column only.

^b Fe and Mo are waterbody specific.

^c Nickel values are also hardness related; values are 25 mg/L when [hardness] < 100 mg/L, and 100 mg/L when [hardness] > 100 mg/L at the site in question.

^d Uranium is calculated as 0.002 [hardness in mg/L] at the site in question.

^e Cobalt objective value to be applied to a dissolved concentration.

Since there are no Saskatchewan or national water quality guidelines for uranium, molybdenum or cobalt, site specific decommissioning water quality objectives were developed based on site specific conditions, the consideration of past, interim, and current guidelines from other jurisdictions, and experimental toxicity data published in the literature.

Water quality decommissioning objectives for flooded pits were established for the upper water columns above an expected chemocline. The objectives take into account their isolation from natural freshwater ecosystems, the potential for colonization by aquatic organisms or occasional use by wildlife, and the low permeability and limited biological activity at the bottom of the pits.

2.3.4.3. Decommissioning radiological objectives

The limits on effective dose to nuclear energy workers under the Radiation Protection Regulations (RPR) are 50 mSv in any year and 100 mSv in any five year period (an average of 20 mSv per year). Given the remaining radiological hazards at the Cluff Lake site, meeting the regulatory limits to protect workers will be straightforward. The objective, therefore, primarily focuses on the application of as low as reasonably achievable (ALARA).

The limit on annual effective dose to a member of the public under the CNSC's RPR is 1 mSv. Pathways analysis was used to verify that exposures to members of the public under a variety of potential land use scenarios are well below this limit both during and after the completion of decommissioning activities.

At the Cluff Lake site, decommissioning activities are expected to reduce radon progeny (RnP) and long lived radioactive dust levels to near background conditions. While ingestion scenarios were also considered in the pathways analysis, the potential exposure to gamma radiation is assumed to be the primary exposure pathway.

Gamma surveys will be conducted at a height of one metre above ground surface in disturbed areas that are potentially contaminated. Areas illustrating average dose rates from gamma exposure in excess of 1 $\mu\text{Sv/h}$ above background (averaged over a 100 m \times 100 m surface, or a 10 000 m² surface), or with a maximum spot dose in excess of 2.5 $\mu\text{Sv/h}$ above background, will be remediated. In most areas, dose rates from gamma exposure are expected to be about 0.1 mSv/h above background, with post-decommissioning dose rates from gamma exposure expected to range from 0.1 to 0.5 $\mu\text{Sv/h}$.

2.3.5. Review of alternatives

A variety of alternative methods of minimizing the environmental effects of potential contaminant sources were identified for each key area of the mine site (e.g. tailings management area, mill area, mine areas).

Once the initial options were identified, contaminant source terms were quantified and a modelling framework was used to identify potential environmental effects. Groundwater and surface water quality modelling was a primary focus of this level of assessment, as long term water quality was identified as the environmental component most likely to be affected by the project. Decommissioning activities were generally expected to have only a minor and short term impact on air quality, ambient radiological levels, geology and terrestrial ecology.

2.3.6. Selection and evaluation of preferred options

After the alternatives and mitigation measures were modelled and assessed against the decommissioning objectives, a preferred option was identified for each area and more detailed source term and contaminant release modelling was completed. The model results were then used as inputs into a pathways analysis to evaluate potential long term ecological effects and potential effects on human health. The assessment incorporated the cumulative effects associated with previous activities (e.g. operational), the proposed project and any additional activities that may influence the site.

This pathways modelling involved calculation of contaminant transport through abiotic (e.g. water, sediment, air) and biotic (e.g. prey items) environmental compartments to predict potential contaminant exposure scenarios for VECs and humans. These exposure scenarios were compared to selected toxicity benchmarks to generate semi-quantitative risk quotients to assist in the prediction of the significance of any potential effects.

Some degradation in groundwater quality in the mining areas is anticipated. Additional effects are also predicted for Island Lake, where effluent discharges from the water treatment systems have resulted in increased concentrations of key contaminants (e.g. uranium, molybdenum and selenium). While these residual contaminants of concern (COCs) may pose a limited risk to non-human biota, the potential adverse effects are not considered significant because they are of limited magnitude, restricted to local populations and reversible, with substantial recovery in the first 50 to 100 years.

While institutional land use controls will be necessary to limit development in the mining areas and the tailings storage areas following decommissioning, the site will be suitable for traditional land uses consisting of

casual access, with trapping, hunting and fishing as the primary sources of site activities.

2.3.7. Follow-up monitoring programme

The CEAA process requires the development and implementation of a follow-up monitoring programme that includes:

- (a) Special investigations to verify the input parameters and key assumptions within the modelling;
- (b) Long term monitoring to ensure that the decommissioning activities are meeting performance objectives and that associated environmental effects have not been, and are not likely to be, exceeded.

The follow-up programme should also include proposed contingency measures if the monitoring indicates that the decommissioning objectives may not be achieved, or that predicted environmental effects may be exceeded.

At Cluff Lake uncertainties in model predictions, including source terms and issues relating to the potential effects of COC on aquatic and terrestrial biota, have been identified and will be the subject of further follow-up during and after the decommissioning. Proposed contingencies have also been identified.

3. CONCLUSIONS

The regulatory framework for decommissioning of uranium mines and mills in Canada has evolved over time. Efforts have been undertaken to harmonize the regulatory process within the various federal jurisdictions and with the provincial regulatory bodies. A systematic process has been developed and used to establish decommissioning objectives based on provincial and national guidelines or, in the absence of guidelines, site specific criteria. These in turn are used to assess the decommissioning options against these objectives and to identify a preferred option. Follow-up programmes can then be developed to confirm predictions or address uncertainties and identify contingency measures when decommissioning objectives are not met. Ultimately, this process will identify a decommissioning approach which will help ensure that potential contaminant sources are removed, minimized and controlled to achieve a stable end state that is safe for both humans and non-human biota over the long term.

REFERENCE

- [1] CANADIAN NUCLEAR SAFETY COMMISSION, Comprehensive Study Report, Cluff Lake Decommissioning Project, CNSC, Ottawa (2003).

Paper 10

REMEDIATION OF THE ZIROVSKI VRH URANIUM MINE

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Abstract

After 25 years of exploration and mine and mill development, production of yellow cake at the Zirovski Vrh uranium mine started in 1984. After only six years of operation production was stopped in 1990. The closure of the state owned mine was neither expected nor planned and there were no funds available for remediation to be carried out in due course. To solve the funding problems the appropriate acts and regulations had to first be adopted by the Parliament of the Republic of Slovenia. The plans for research, development and remediation design and a financial plan were prepared by the mine company and contractors. Additional permits for the remediation of the underground mine, waste piles, and mill tailings were obtained. The legislative problems were solved by the new Act on Ionising Radiation Protection and Nuclear Safety, adopted by the Slovenian Parliament in 2002. The Government of the Republic of Slovenia raised a loan from the European Investment Bank to enable implementation of the remediation plans. The milling facilities were demolished, contaminated areas decontaminated and 90 000 t of debris and contaminated soil were stored on the mine waste pile. Field work is continuing in the underground mine, as is the remediation of surface objects. The required design data for the long term remediation of the mill tailings (the base of the tailings affected by a landslide, tailings stabilization, and the cover system) are under intensive preparation. The temporary mine waste piles are being relocated to the central mine waste pile for disposal. The total costs of the remediation will be €37.3 million. The state budget will contribute the project management and the operating costs of €13.4 million and a European Investment Bank loan will cover the investment costs of €23.9 million. Recontouring and placement of a cover on the mine waste pile will start in the second half of 2004. Field work on the mill tailings will start in 2005. It is planned that all field work will be finished by the end of 2006.

1. INTRODUCTION

The Zirovski Vrh uranium mine (Rudnik Zirovski Vrh, RZV) is located 45 km west of Ljubljana. The uranium mineralization in Groeden sandstone was discovered in 1960 and ore production started in 1982. The ore processing plant started in 1984. Only 610 000 t of ore with a uranium content of 0.7 kg per

tonne of ore was processed, resulting in a production of 452 t of yellow cake during the six years the mill operated. Production ceased in 1990 after a decision of the Slovenian Government, with no notice given to the operator.

The mine and its facilities lie on the north-eastern slopes of the Zirovski Vrh ridge (960 m). The region belongs to the foothills of the Julian Alps. The elevation of the underground mine ranges from 430 m (bottom of the river valley) to 580 m (P-1 adit). The mill was located in the valley of the Brebovsica river (415 m) and the mill tailings are located on the southern slope (530–560 m) of the Crna Gora hill (611 m). A total of 3 307 000 t of material was excavated (633 000 t of high grade ore, 206 000 t of low grade ore, and 2 468 000 t of spoils).

The mine is located in a populated farming area. The radioactive impact is small, the yearly dose contribution ranging from 0.3 to 0.4 mSv/a [1]. Uranium and radium concentrations in nearby surface water flows are elevated.

2. GENERAL REMEDIATION OBJECTIVES

For emissions of contaminants via the water and air pathways the following limits were set by a decision of the Ministry of Health of the Republic of Slovenia and are shown in Table 1 [2].

Non-radiological requirements for the discharge of contaminants in water are defined by the environmental legislation on the conservation of water resources. Additionally, local permits define the basic conditions for the operation of the mining and milling sites and also for their closeout. In the case of the P-1 and P-9 waste piles, the local permit allows only temporary storage of the mine waste. The requirements stipulated in the local permits are taken into consideration in RZV's remediation plans.

3. UNDERGROUND MINE WORKINGS

3.1. Description

The mine workings extend 2000 m in a north–south direction, 150 m east–west and over a height of 180 m. About 60 km of mine openings were constructed, mostly having a diameter large enough to allow transport and production. The mine is divided into four galleries, the galleries in turn into 14 blocks 200 m wide. Uranium ore was mined by the room and pillar method.

TABLE 1. AUTHORIZED LIMITS ISSUED FOR THE CLOSEOUT OF THE LIABILITIES OF THE ZIROVSKI VRH MINE [2]

Location	Contaminant	Concentration	Unit	Annual load	Unit
Discharge water					
Mine (P-10)	Uranium	250	mg/m ³	170	kg
	²²⁶ Ra	60	Bq/m ³	50	MBq
Jazbec	Uranium	510	mg/m ³	85	kg
	²²⁶ Ra	40	Bq/m ³	25	MBq
	²³⁰ Th, ²¹⁰ Pb, ²¹⁰ Po (total)	100	Bq/m ³		
Boršt	²²⁶ Ra (annual average)	60	Bq/m ³	50	MBq
Airborne					
Mine, Jazbec	²²² Rn	0.1	Bq/m ² ·s		
Borst	²²² Rn	0.74	Bq/m ² ·s		
External radiation	Average over the first 1.25 m above ground	200	nGy/h		
RZV (total)	Annual effective dose	0.3	mSv/a		

The underground mine workings are connected to the surface by shafts and adits that were used for ventilation, material supply and ore transport. Adits P-1 and P-36 are used mainly for ventilation. The ventilation of the underground mine is effected by a slight vacuum, induced by the ventilation stations at these adits. Mine air is removed while fresh air flows in via the ventilation shafts and adits distributed over the entire area of the mine. The main mine access is through adit P-11. The deepest adit, P-10, is used for mine water discharge.

In the north-western part the uranium bearing structures of Zirovski Vrh are overthrust by the Jazbec scale (Groeden rocks). It is estimated that this thrust represents an impermeable barrier with a hydraulic conductivity of 10^{-9} m/s. The overlying layers of sandstone and conglomerates have a high hydraulic conductivity [3].

Water inflows into the mine mainly occur along fractures and fault zones in the sandstone and conglomerates. The conglomerates with a carbonate matrix represent a particularly important aquifer (3rd horizon). The north-western part of the mine is more permeable because of the fractures.

Groundwater entering the mine becomes contaminated by passing through ore zones that are intersected by the faults. Contaminated water is

drained into open mine workings and exploration boreholes that were drilled from the tunnels. Stopes that were constructed below the levels of the saturated zone are flooded. The water in these tunnels has uranium concentrations exceeding 6 mg/L. During ore production the inflow of groundwater in the upper parts of the mine decreased. The highest inflow of water is presently found on the deepest mine levels.

The yearly average uranium concentration in the mine water discharged through adit P-10 ranged between 250 and 350 $\mu\text{g U/L}$ in the period from 1992 to 2000. The ^{226}Ra concentration was between 30 and 100 Bq/m^3 during the same period. The total discharge from the mine ranged between 15 and 30 L/s [3]. A decrease of uranium and radium concentration in the mine water was observed after uranium production ceased. Iron concentration in the discharged mine water is below the detection limit. The sulphate concentration is very low compared to discharge concentrations observed in other mines.

3.1.1. Objectives of mine remediation

The objectives regarding the remediation of the mine as defined in Ref. [4] are:

- (a) Reduction of radon emission from the underground mine openings;
- (b) Long term stabilization of underground mine workings to prevent the inflow of surface waters to the mine that result from the collapse of mine openings;
- (c) Permanent discharge of mine water through adit P-10 meets the regulatory limits for uranium;
- (d) Permanent discharge of uncontaminated groundwater through P-11 and P-9;
- (e) Protection of surface and groundwater (within the regulated limits);
- (f) Reclamation of the surface area in the surroundings of shafts, raises and adits,
- (g) Prevention of unauthorized access to underground openings.

For better collection of the inflowing groundwater and to avoid contact of groundwaters with ores, drainage boreholes between 150 and 170 m long have been drilled in the deepest part of the mine since 1993; the uranium concentration in the inflowing groundwater decreased simultaneously.

Adit P-10 was completely remediated over a length of 520 metres (out of a total length of 1200 metres) between 1996 and 1997. Further mine facilities on the surface, e.g. the crushing plant, the chemical plant and transport bridges, were torn down or handed over to other users after decontamination.

4. WASTE ROCK PILES

4.1. Description of the mine waste rock piles

Minor waste rock piles P-1 (70 000 t) and P-9 (143 000 t) are situated close to adits P-1 and P-9, respectively. Jazbec is RZV's main waste rock pile (1.5 million t) and located in the Jazbec valley right below the entrance to adit P-11. The slope of the waste pile is 1:2 with 3 m wide berms placed at 12 m height intervals. At present the highest point of the waste pile is 493 m above sea level. On the supporting dam, formed by mine waste on the eastern pile face, a road leads to the former crusher site. Judging by the water levels measured in the waste pile the dam is less permeable than the stored material. No seepage water is observed on the pile face.

In addition to waste rock, red mud from uranium ore processing was disposed off on the waste pile. The red mud is a precipitate of the uranium leaching solution produced by neutralization with lime. The main components are calcium, gypsum, iron hydroxide, uranium (0.5 Bq/g), ^{226}Ra (0.2 Bq/g) and ^{230}Th (62 Bq/g). The ^{230}Th concentrations in the tailings are enriched by a factor of eight compared to the ^{226}Ra concentration. The material was stored in alternating layers of 80 cm of mine waste and 20 cm of red mud. A 5 m thick zone on the surface of the pile consists of coarse mine waste only. Demolition debris from the decommissioning of the milling site was deposited on the waste pile as well.

4.2. Hydraulic regime

There are no water sources in the area of waste pile P-1. About 0.5 L/s of uncontaminated mine water is discharged from adit P-9 and passes waste pile P-9. The uranium concentration of this water is about 400 $\mu\text{g/L}$.

The Jazbec valley is cut by the Hotavlje fault, with some wet moor areas on the surface. At the bottom of the Jazbec valley are dark grey limestone outcrops showing characteristic karstic properties. Along the Brebovščica river numerous springs exist that are fed by karstic groundwater. There is a 329 m long concrete channel at the bottom of the waste pile. In the upstream section (152 m) the channel has a diameter of 1 m and in the downstream section (177 m) 3 m. This channel collects the water from three creeks in the upstream valleys as well as the drainage water from the pile. This water is discharged into the Brebovščica River. Before construction of the waste pile a drainage system was built at the bottom of the valley. Small springs were diverted by pipes connected to the concrete channel. At present the drainage channel is in good condition; however, the drainage pipes are mostly in a poor state. Currently,

additional horizontal drainage boreholes are being drilled from a large diameter well in the upper part of the waste pile (near P-11) to reduce groundwater inflow to the pile from the hinterland.

The groundwater is contaminated because of the infiltration of meteoric water that percolates through the uncovered waste pile and red mud. Various sampling points on and around the waste pile allow evaluation of the drainage water quality. The water at the bottom of the waste pile contains up to 5 mg/L of uranium. Downstream of the pile, in the alluvial terrace of the Brebovščica River, no groundwater is found (borehole BS-20), while concentrations of about 150 $\mu\text{g/L}$ uranium were measured in the deeper karst aquifer. The Brebovščica River springs downstream are probably fed by water from the karstic limestone layer [3].

The water discharged from the drainage channel at the bottom of the waste pile has an annual average uranium concentration in the range of 250 to 550 $\mu\text{g/L}$ (1991–1999). In the same period the sulphate concentrations were between 200 and 300 mg/L, which is about 10 times higher than in the mine drainage.

4.3. Objectives of mine waste remediation at Jazbec

The specific aims of the technical measures for the closeout of the mine waste pile are:

- (a) Limitation of the radon exhalation;
- (b) Limitation of leaching of contaminants and erosion protection of the mine waste pile by covering;
- (c) Limitation of the effect of surface and groundwater from the hinterland on the mine waste (erosion, wetting of waste);
- (d) Ensuring the geomechanical stability of the mine waste pile, namely erosion resistance, by reshaping the mine waste pile;
- (e) Prevention of erosion and excessive wetting of the cover by means of a drainage system;
- (f) Relocation of the waste rock piles P-1 and P-9 to the Jazbec mine waste disposal site.

5. THE BORŠT MILL TAILINGS DISPOSAL SITE

5.1. Description of the mill tailings

From the beginning of operation until the end of uranium production at RZV, 600 000 t of hydro-metallurgical tailings had been generated and deposited at the Boršt tailings pile. The volume of the deposited material is approximately 375 000 m³. The tailings pile covers an area of 4.11 ha and is about 2 km away from the former uranium processing plant. There is an access road at the southern and western periphery of the pile. To allow access to the tailings pile with heavy machinery, regularly spaced roads were constructed on the tailings pile (at every 5 m of elevation) using 73 000 t of mine waste rock. The tailings are deposited on a natural slope.

During deposition of the tailings the following measures were taken concerning the environmental controls, water management and geotechnical stability of the tailings:

- Diversion of surface waters out of the tailings pile to prevent contact with tailings;
- Capturing of groundwater and springs and their linkage to the drainage system;
- Sealing of the bottom of the tailings pile using clay material;
- Capturing of (contaminated) surface and seepage water from the tailings and discharge into the retention pond;
- 20° embankment slopes with horizontal berms.

After the planned elevation was reached, the tailings surface was covered with material that had previously been removed from the base of the tailings pile and re-vegetated with grass. The general slope of the tailings pile at the northern part is approximately 20°, the inclination between berms is 24–27°. The slope was covered with 25 cm of topsoil and vegetated with grass. The top plateau of the tailings pile is partly covered with waste rock from the construction of the drainage tunnel below the tailings pile. The provisional covering was done to reduce the radon exhalation.

The tailings are a relatively coarse ground sandy-aleuritic residue of the uranium ore processing with a grain size of less than 0.5 mm, but with more than 50% larger than 0.125 mm. The average uranium content is 0.9 Bq/g or 80 g/t U₃O₈ and 8.6 Bq/g of ²²⁶Ra. They mainly consist of SiO₂, calcite and sulphate salts. The tailings were deposited on site with a water content of 20–24%. Presently, the tailings are wet and tixotropic, referring to the latest geo-mechanical investigations. According to the planning documents there should

be no danger of liquefaction. However, during deposition liquefaction of the material was observed.

Before the placement of tailings started, the ground for the base of the pile was prepared using the material removed during the site preparation work. Drains and pipes were laid on the surface and covered with a layer of clay. Water from two springs that were to be buried by the tailings was captured and drained in two PVC pipes. The discharge water from these springs is sampled at the sampling point SDIJ outside of the tailings pile.

Due to the high radium content in the tailings, both the uncovered surface area and the covered embankments are a considerable source of radon. The exhalation was measured to be around $5 \text{ Bq}/(\text{m}^2\cdot\text{s})$ (with a range between 1 and $10 \text{ Bq}/(\text{m}^2\cdot\text{s})$). Surface waters, as well as water in tailings and underneath, are contaminated by uranium and soluble inorganics (NH_4^+ , SO_4^{2-} , Cl^-). Ammonia and total dissolved salts are above the regulated discharge limits.

The geological foundation of the Boršt tailings pile consists of Upper Triassic (Carnian) sediments, yellow, red and grey clay, alternating with aleuritic layers with tuff and tuffaceous inclusions. A large number of sub-vertical faults disrupt the geological base.

After heavy rainfalls in November 1990 a crack with 20–30 cm of vertical movement appeared in the road on top of the tailings due to a landslide. The extent and depth of the sliding area have been established by surveying, drilling and borehole inclination measurements. The landslide occurred on the contact between the Carnian clastites and the tuffaceous rocks. An evaluation of the borehole survey confirmed that the tailings pile had been placed on an old landslide (paleoslide). The landslide underlies the major part of the Boršt tailings pile and involves a large part of the geological foundation. The general movement of the slide was towards the north. The average thickness of the slide is 50 m.

Tailings moved by landslide:	328 579 m^3
Masses of ground moved by landslide:	2 593 175 m^3
Total volume of the landslide:	2 921 754 m^3

Construction of the drainage tunnel in 1995 had a significant checking effect on the landslide. The tunnel starts in the sliding face, crossing the main sliding plane at about 214 m from the entrance of the tunnel. At the end of the tunnel two drainage wings were excavated and where a drainage screen is connected to 21 boreholes drilled from the surface. The distance between each borehole is about 10 m. The inflow into the drainage tunnel averages around 1.3 L/s (40 000–60 000 m^3/a). The drainage boreholes contribute 40% of the

inflow, while 60% enters through the tunnel face. Construction of the tunnel has a considerable effect on the local groundwater regime. Most of the water levels measured in the piezometers in the tailings pile and in the close vicinity of the site decreased. However, no correlation to the distance from the drainage tunnel was found. After construction of the tunnel the discharge of the captured springs measured at SDIJ rapidly decreased to about 20%.

5.2. Objectives of the Boršt mill tailings remediation

The remedial objectives concerning the tailings are to:

- (a) Permanently stabilize the landslide by a drainage channel, drainage screen, and cover construction;
- (b) Stabilize the tailings and enhance the erosion resistance by recontouring and covering the tailings pile;
- (c) Prevent radon exhalation, the leakage of hazardous contaminants into surface and groundwaters;
- (d) Protect the tailings pile from the surface and groundwaters from the hinterland (erosion, increased infiltration through the cover and the tailings);
- (e) Prevent erosion of the cover and the tailings by construction of a drainage system.

5.3. Investment costs and funding

When the last closeout programme was submitted to the Government, cost estimates were prepared for both fixed and current prices due to inflation (in Euros and in Slovenian Tolar). In the following, the presentation of investment costs and estimated fund sources of the project are presented as fixed prices in Euros [5].

Closeout of the Zirovski Vrh uranium ore mine has been in progress since 1990. Costs incurred to the end of the year 2000 were estimated on the basis of the actual expenditure up to the time when the revision to the programme and plan for funding estimated the total expenditure at €29.4 million. Expenditures in the year 2000 were about €2 million.

According to the revision of the programme that the Government of Slovenia adopted in the year 2001, future costs of the project implementation will amount to €37.3 million (including VAT) over a period of five years. As the project actually started at the beginning of the year 2002, we estimate that it will be completed by the end of 2006.

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Total closeout costs (including VAT) will amount to €68.7 million for the entire period between the years 1990 and 2007. These investment costs do not include the following cost items:

- (a) Costs for redundant employees and unforeseeable costs.
- (b) After the closeout a 5 year environmental monitoring programme was planned. The annual costs were estimated at €127 000 (in total €633 000), but were not incorporated into the total closeout costs.
- (c) The costs of liquidating RZV have not been explicitly included.

The total costs of closeout arising from 1 January 2002 to the final closeout amount to €37.3 million and are detailed in Table 2 [6].

Other cost elements include:

- (1) Costs of safety at work;
- (2) Operating expenditures for, e.g. the laboratory, operation and maintenance of various plants, etc.;
- (3) Material and services related to the operation of RZV, contributions to the community for new developments, various types of contracts, and training;

TABLE 2. SUMMARY OF THE REQUIRED COSTS AND SOURCES OF FUNDING OF THE PROJECT (Millions of €)

	Total 2002–2006	2002	2003	2004	2005	2006
Cost element						
Ore mining	16 438	3 660	5 883	3 837	1 451	1 607
Concentrate production	613	132	0	0	0	481
Waste pile remediation	6 829	0	185	3 352	1 843	1 448
Other	8 863	1 797	1 936	1 814	1 714	1 602
Total	32 742	5 457	8 136	9 003	5 008	5 183
VAT	4 558					
Total	37 300					
Source of funding						
Government allocation	17 300					
European Investment Bank (EIB) loan	20 000					

- (4) Labour costs for 36 employees;
- (5) Indemnification that may need to be paid to land owners in case of possible intervention on their land;
- (6) Purchase of equipment (e.g. for decontamination of motor cars, tools and devices during the implementation phase).

In the past the closeout of the Zirovski Vrh uranium mine was financed from Slovenian Government contributions and its own income from, e.g. the hiring out of employees and the lease or sale of fixed assets.

It was found that in the past the budgetary sources were not sufficient to carry out the closeout programme on schedule. Their irregular inflow made the work even more difficult, but Zirovski Vrh solved its liquidity problems by generating its own income.

According to the revision to the programme, and as envisioned by the Government, for the future closeout work three types of funding sources are expected: the original budget sources, EIB loans and income generated by the company itself. The EIB loans will be used for the mine closeout work and for mine waste pile and mill tailings remediation; the money coming directly from the Government budget will cover other costs and VAT; the company's revenue will be used for funding the layoff of employees, a cost item that is not incorporated in the estimated amount of investment costs (see Table 2).

The budgeting process is completely governed by the annual allocation of funds in the government budget. The volume of funding is developed by the Ministry of Environment and Spatial Planning (MoESP) in consultation with other governmental entities such as the Ministry of Finance, Ministry of Economics, etc., and on the basis of the information submitted by RZV. The resources are targeted and the basic assumptions for the fiscal year are provided by the MoESP. RZV prioritizes the work by adjusting the yearly scope of work to the funding available.

6. SUMMARY AND CONCLUSIONS

Following 25 years of exploration and mine and mill development, production of yellow cake at the Zirovski Vrh uranium mine started in 1984. After only six years of operation production was stopped in 1990. The closure of the state owned mine was neither expected nor planned and there were no funds available for the remediation to be carried out in due course.

To solve the funding problems, the appropriate acts and regulations were adopted by the Parliament of the Republic of Slovenia. The plans for research, development and remediation design and the financial programme were

prepared by the mining company and contractors. The supplemental location permit for the remediation of the underground mine, waste piles and mill tailings were obtained. The legislative problems were solved by the new Act on Ionizing Radiation Protection and Nuclear Safety, adopted by Parliament in 2002. The Government of Slovenia raised a loan from the EIB to enable the implementation of the remediation plans.

The milling facilities were demolished, contaminated areas decontaminated, and 90 000 t of debris and contaminated soil were disposed of on the mine waste pile. The fieldwork is going on in the underground mine, as is the remediation of the surface facilities. The required data for designing the long term remediation of the mill tailings (i.e. dealing with the landslide at the base of the tailings, the tailings stabilization and the cover system) are under intensive preparation. The temporary mine waste piles are being relocated to the central mine waste pile for final disposal.

The total costs of the remediation project will be €37.3 million. The State budget will contribute the project management and the operating costs of €13.4 million and the EIB loan will cover the investment costs of €23.9 million. Reshaping and cover emplacement at the mine waste pile were to have started in the second half of 2004. Mill tailings field work will start in 2005. All field work is planned to be finished by the end of 2006.

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Paper 11

ENVIRONMENTAL REMEDIATION OF URANIUM PRODUCTION FACILITIES IN SPAIN

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Abstract

Beginning in 1973, ENUSA carried out uranium mining and milling activities in the areas of Saelices el Chico (Salamanca) and La Haba (Badajoz). The total amount of uranium produced was 6000 t U_3O_8 . The uranium facilities at La Haba were closed and remediated in the years from 1991 to 1997, followed by a five year (1998–2003) supervising programme to verify compliance with remediation design criteria (groundwater quality, erosion control, radon and gamma radiation exposure). The uranium facilities at Saelices el Chico were phased out at the end of 2000 and the current activities are the decontamination, dismantling and decommissioning of the ore processing facilities (Elefante and Quercus plants) and remediation of the open pit mine. Also the decommissioning of the waste rock disposal, heap leaching pad and tailings dam are part of the environmental remediation programme (2000–2008). In the paper, ENUSA presents information on the dismantling and decommissioning work, waste management and water treatment, representative remediation costs in Spain and other relevant aspects of these environmental remediation activities.

1. CHRONOLOGY AND SCHEDULE OF REMEDIATION ACTIVITIES

Uranium mining operations in Spain were developed during the second half of the last century (Fig. 1), mainly to supply raw materials for nuclear electricity generation that in some years accounted up to 30% of the total electricity market.

In the 1990s, because of the low price of uranium, the phaseout period began, resulting in the dismantling and decommissioning of the uranium production facilities according to the schedule shown in Fig. 2.

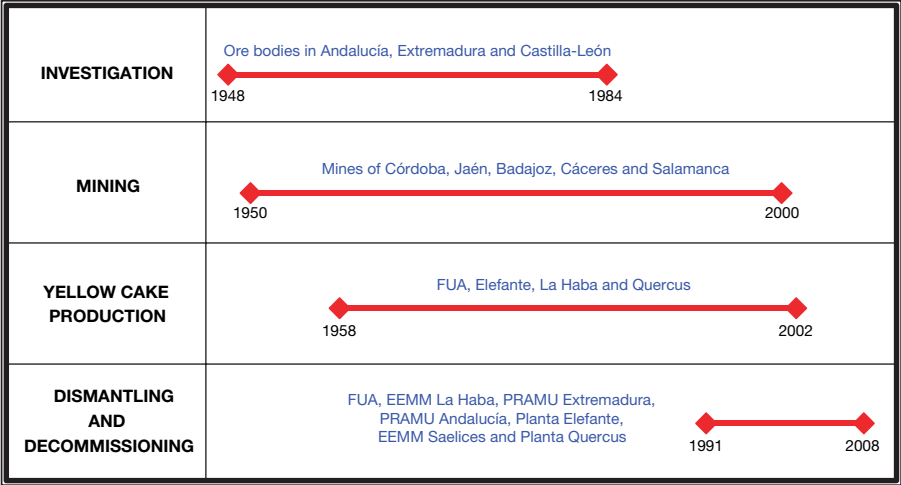


FIG. 1. Chronology of uranium mining in Spain.

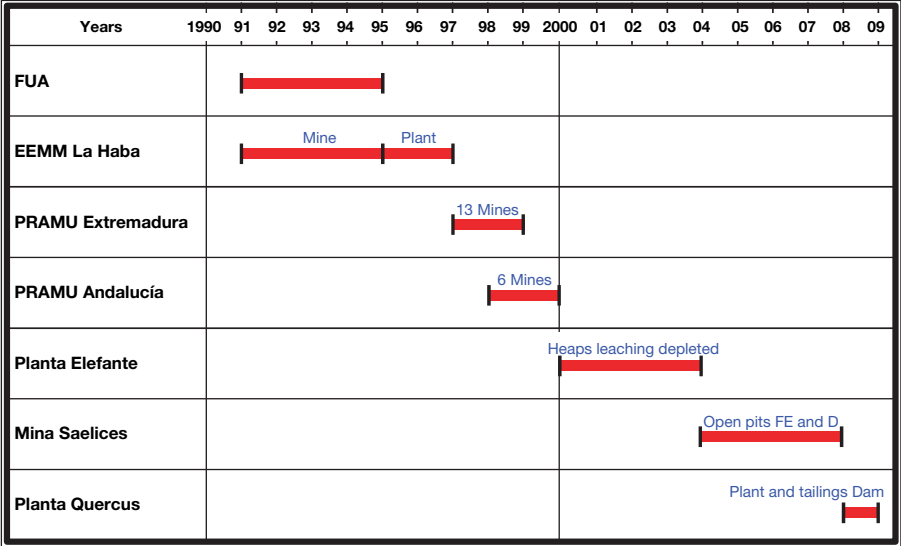


FIG. 2. Schedule of phaseout of uranium production facilities in Spain.

2. DECOMMISSIONING REGULATIONS

Spanish regulations require that the facility owner, prior to finishing production activities, present a dismantling programme to the Ministry of Economy (MINECO). This programme is to be evaluated by the Nuclear Safety Council before authorization is granted to the owner to undertake dismantling and decommissioning operations.

The dismantling programme comprises the following steps:

- (a) Safety analysis:
 - (i) Operational history of the facility describing the facility and the processes, as well as an inventory of installations, waste rock and tailings to be dealt with during the decommissioning;
 - (ii) Decommissioning criteria, analysing the various alternatives considered and corrective actions to be implemented with respect to land decontamination, materials declassification, protection from barrier degradation and human intrusion;
 - (iii) Radiological site characterization, paying special attention to natural background radiation of the area, which will serve as a reference for achieving decommissioning objectives; evaluation of characteristic parameters related to meteorology, hydrology, hydrogeology, water and soil use, geology, seismicity and geotechnology, demography, as well as socioeconomic factors;
 - (iv) Analysis of the radiological impact on workers and the public for both the operation of the decommissioning phase, as well as the final impact on the public; once decommissioning is complete, radiation sources and different exposure pathways are evaluated;
 - (v) Post-decommissioning monitoring programme of the radiological conditions at the decommissioned site, to be implemented during a final verification period (minimum five years/maximum ten).
- (b) Operating procedures, paying special attention to organization and rules to be implemented under normal conditions and in cases of accidents;
- (c) Technical specifications applicable during the dismantling;
- (d) Quality assurance programme, ensuring that operations are carried out in compliance with established criteria; these procedures include aspects such as organization, control and design, verification, operating procedures and instructions, document and archive control, inspections and audits;
- (e) Radiological protection programme;
- (f) Emergency plan (internal);
- (g) Radioactive waste management plan;

- (h) Site remediation, in which the technical basis for the design, monitoring and treatment of liquid wastes, solid materials and waste management are established, as well as the design of covers and stability analysis of the decommissioned structures and final remediation of affected areas.

3. HISTORICAL INFORMATION ON THE REMEDIATION ACTIVITIES AT LA HABA (BADAJOZ)

3.1. Site characteristics

The uranium mining and milling facilities were closed and remediated by ENUSA (1991–1997). This section presents the more relevant aspects of this environmental rehabilitation.

The main properties of the site are outlined in the following:

- (a) Topography: hills with gentle slopes;
- (b) Geology: Ordovician-Silurian metasediment in contact with granite;
- (c) Hydrogeology: deep water in faults and fractures;
- (d) Climate type: Mediterranean subtropical with a dry and warm summer; mean annual precipitation: 450 mm; annual evaporation rate: 1300 mm;
- (e) Demography: average population density of the district <9.5 inhabitants/km²; nearest population: 10 000 m;
- (f) Materials: waste rock and overburden have a high clay content that make them suitable for backfilling and ground sealing.

3.2. Description of the facilities

The main features are listed in the following:

- (a) Mine (El Pedregal): open pit mine (surface area 20 ha); waste rock piles: 8×10^6 t (surface area 20 ha);
- (b) Plant (Lobo-G): heap Leaching piles: 3.5×10^5 t (surface area 5 ha); experimental milling plant: 32 t (U₃O₈)/a (crushing, leaching, counter current washing, resin in pulp, solvent extraction and precipitation, drying and packaging);
- (c) Tailings dam: capacity: 1×10^5 /m³ (surface area: 4 ha) [1]; slopes: 2.5:1 (upstream), 3:1 (downstream); Antiseismic design: up to 0.15 g (horizontal) and 0.10 g (vertical);
- (d) Water treatment; Natural and forced evaporation.

3.3. Environmental and radiological protection criteria

The following criteria and requirements have been put forward:

- (a) The flow of radon exhalation from soils after the end of operations should be lower than $1 \text{ Bq/m}^{-2}/\text{s}^{-1}$. This value is close to the natural background of the area.
- (b) The gamma exposure rate, measured at 1 m above the surface of the land, should be less than 0.20 mGy/h above the natural background of the area. The value of the natural background is 0.23 mGy/h .
- (c) The effective equivalent dose to the public should be lower than 1 mSv/a .
- (d) The groundwater activity concentration will be lower than the notification limits stipulated in Ref. [2].
- (e) The concentration of residual activity in soils should not be more than 1 Bq/g above the value of the natural background when later used as pastures or forest.
- (f) The design of the cover should ensure the stability of the structure for at least 200 years.

3.4. Decommissioning/closeout plan

The decommissioning and closeout plan comprises the following elements:

- (a) Natural radiation background assessment;
- (b) Characterization of the materials;
- (c) Backfilling of the open pit mine;
- (d) Regrading of slopes in waste rock piles;
- (e) Dismantling and demolition of the plant;
- (f) Stabilization of the tailings pond;
- (g) Landscape integration.

3.5. Land use after remediation

Taking into account the different radiological exposure pathways, the distribution of population and the present use of the surrounding land, possible future land use scenarios were analysed (see Table 1) and ENUSA proposed use as forestry and pasture to the regulatory authorities.

TABLE 1. ASSUMED RESIDUAL RADIONUCLIDE ACTIVITIES AND RESULTING DOSES

Land use scenario	Residual radioactivity in soil (Bq/g)	Resulting doses (mSv/a)
Industrial	1.6	1.07
Forestry	6	0.99
Dairy (adult)	2	1.04
Dairy (young)	2	0.89
Agriculture	1	1.12

3.6. Environmental monitoring

Using as reference the environmental monitoring undertaken during the operations of facilities a long term sampling programme was designed, as shown in Table 2.

3.7. Costs and funding

There is an important reduction in main cost items when remediation activities are undertaken by the former operator of facilities, as shown in Table 3.

The financing of the remediation work was shared by ENRESA and ENUSA, because ENRESA has the liability for decommissioning the old mines.

4. PRESENT AND ONGOING RESTORATION ACTIVITIES AT SAELICES (SALAMANCA)

4.1. Description of the facilities

At the end of the year 2000 the uranium ore processing activities were phased out and the period of decontamination began with dismantling and decommissioning of mines and ore processing facilities ('Elefante' and 'Quercus' plants).

- (a) Mine: Open pit mine (surface area: 200 ha); waste rock piles: 68×10^6 t (surface area: 150 ha); pending the abandonment of the licence;

TABLE 2. SAMPLING PROGRAMME

Type of sample	Sample collection				Sample analysis	
	No. of samples	Location	Method	Frequency	Frequency	Method
Air	1	La Haba	Continuous	Monthly filter change	Monthly	Gross α
	6	Facilities	Passive dosimeter	Semi-annual dosimeter change	Quarterly Semi-annually	U_N , ^{230}Th ^{226}Ra , ^{210}Pb , ^{222}Rn
	7	Outside facilities	Grab	Annually	Annually	Rn-flux
Water	8	Creeks	Bailing	Quarterly	Quarterly	Gross α/β , U_N , ^{230}Th
	19	Wells/ boreholes	Pumping	Monthly	Monthly	Gross α , U_N , ^{226}Ra , ^{210}Pb ,
				Quarterly	Quarterly	Gross β , ^{230}Th , ^{226}Ra , ^{210}Pb
Direct radiation	6		TLD	Quarterly	Quarterly	γ exposure rate

TABLE 3. PROJECT COST ITEMS

Items	Cost	Remarks
Planning, engineering and licensing	€5 × 10 ⁵ (total cost)	Environmental impact statement Dismantling project Natural background evaluation Radiometric study Safety analysis
Tailings ponds	€1 × 10 ⁶ (total cost)	Volume = 1.5–105 m ³
Waste rock piles:		
In situ stabilization	€0.5 /t	Mass = 8 × 10 ⁶ t
Transport and backfilling open pit	€1.0 /t	Surface area = 20 ha
Plant disassembly and soil cleanup	€5 × 10 ⁵ (total cost)	Experimental plant (heap and dynamic leaching) Capacity = 500 t/d
Mining facilities	€3 × 10 ⁶ (total cost)	Open pit surface = 25 ha Capacity = 3000 t/d
Remediation and revegetation of reclaimed areas	€1.0/m ²	Surface = 60 ha
Forced evaporation of liquid effluents	€1.5/m ²	Volume = 4105 m ³
Post-decommissioning surveillance programme	€2 × 10 ⁵ /a	Over five years

- (b) Elefante plant: heap leaching piles: 7.2106 t (surface: 27 ha); solvent extraction and precipitation, drying and packaging plant; dikes with neutralization slurry: 0.3 × 10⁶ t (surface area: 17 ha); decommissioning nearly completed;
- (c) Quercus Plant: crushing, sorting, leaching, counter current washing, solvent extraction and precipitation, drying and packaging; heap leaching piles: 1 × 10⁶ t (surface area: 8 ha); tailings dam: 1 × 10⁶ t (surface area: 20 ha); water treatment facility: 75 m³/h; the decommissioning project will be presented for licensing at the end of 2004.

4.2. Decommissioning of the ‘Elefante’ plant

The decommissioning of the Elefante heap leaching plant comprised the following elements:

- (a) Dismantling and demolition of the plant;
- (b) Decommissioning ‘in situ’ of heap leaching piles;
- (c) Regrading of the slopes to a stable ratio of 1:5;
- (d) Multilayer capping with a 2.3 m thick cover consisting of 0.9 m of clay, 0.9 m of waste rock, and of 0.5 m of topsoil;
- (e) Stabilization of the slurry dikes.

The decommissioning operations were to have been finished late in 2004.

4.3. Dismantling of the ‘Quercus’ plant

Uranium ore mining and processing were phased out during the year 2000. Until the year 2008 the only operation will be the treatment of mine waters and seepage from the heap leaching pile. The heap leaching pile will be transported to the tailings dam. From now until 2008 the tailings dam will be dewatered and at the end of that year closed out using multilayer capping. At the end of 2007 ENUSA will take the decision of ‘cleanup and mothballing’ versus ‘dismantling and demolition’ of the plant.

4.4. Closure of the open pit mines

The decommissioning of the open pit mines will comprise the following elements:

- (a) Backfilling of the mining area and recontouring to the former topography using the material from the waste rock piles;
- (b) Construction of channels and pond to collect the contaminated water from the catchment area;
- (c) Treatment of the contaminated water;
- (d) Capping the surface with clay, waste rock and soil;
- (e) Revegetation.

4.5. Cost and funding

The total cost of decommissioning the plants and closure of open pit mines will be close to €70 million and the financing will be shared by ENUSA and ENRESA.

5. CONCLUSIONS

- (a) After fifteen years of remediation of uranium facilities in Spain, today a regulatory framework exists that is realistic and suitable for licensing of these activities;
- (b) The know-how of the companies involved in environmental remediation is a guarantee to achieve the final goals of remediation, that is land use with few post-decommissioning restrictions, except for tailings ponds;
- (c) At the end of the decommissioning work the remaining radiological exposure in the area is not significant compared to the natural background at uranium ore sites;
- (d) The funding of the remediation work in Spain is guaranteed because ENRESA has the liability for decommissioning the old uranium mines;
- (e) At present there is a regulatory gap concerning the liability associated with the long term institutional control after decommissioning work.

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Paper 12

URANIUM MINING AND REMEDIATION OF THE STRÁŽ DEPOSIT IN THE CZECH REPUBLIC

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Abstract

The second half of the twentieth century saw large scale development in the exploration and production of uranium ores in the Czech Republic. Many uranium deposits were discovered on its territory. Over a period of 50 years, approximately 110 000 tonnes of uranium concentrate were produced. Different mining methods were developed and used. The extensive production of uranium led to widespread environmental impacts and contamination of groundwaters. After 1990 a large scale environmental programme was set up. The costs of all the remediation activities are expected to be in excess of €25 billion.

1. INTRODUCTION

The history of uranium exploitation in the Czech Republic (and in the former Czechoslovakia) dates back more than 55 years. Over the initial period, from 1946 until the beginning of the 1950s, exploitation was mainly carried out in the reopened mines of the Jáchymov mining area. The rapid development of surveying and extraction work was reflected in the great growth of exploitation in other areas of Bohemia and Moravia, involving the regions of Příbram, of Hamr-Stráž pod Ralskem and Dolní Rožínky, i.e. southern and western Bohemia (Fig. 1). More than 100 000 tonnes of uranium have been extracted from over 800 trial and production shafts since 1946.

Owing to the diversity of the deposits, uranium exploitation was carried out with the whole spectrum of mining methods available, which were selected as appropriate for the host rock at a given locality. Two basic methods of uranium extraction are applied in the Czech Republic:

- (1) Conventional underground mine workings;
- (2) Underground leaching in situ.



FIG. 1. Location of the Hamr-Stráž pod Ralskem deposit in the Czech Republic.

2. DISCOVERY AND SURVEY OF THE DEPOSITS

The deposits in the area of Hamr-Stráž were discovered in the 1960s. In 1963 an aerial geophysical survey detected high magnetic anomalies into which a borehole, HJ-1 (Hamr na Jezeře-1) was drilled. Following the detection of the anomaly at well HJ-1, other exploration boreholes were drilled in its vicinity and all of them confirmed uranium mineralization. Well HJ-1, located in the Hamr deposit, represented the beginning of an exploration borehole network.

3. UNDERGROUND EXPLOITATION

Underground exploitation of uranium ore has been the main method used throughout the existence of the uranium industry in the Czech Republic (and especially during Czechoslovakian times). Underground exploitation proceeded using a whole spectrum of mining methods, selected according to the types of deposits at the given locality. Both classic mining methods and alternatives were employed:

First type: vein deposits — exploited employing mainly the ‘overhand stoping’ method in various variants;

Second type: zone deposits — exploited using ‘overhand stoping’ and ‘underhand stoping’ methods in various variants;

Third type: deposits in tectonic zones — exploited employing various variants of the ‘open room, cave-in’ or ‘overhand stoping’ method;

Fourth type: stratiform deposits in sediments — exploited using mainly the ‘longwall mining’ method or transversal incisions;

Fifth type: sedimentary deposits of the Northern Bohemian Cretaceous — exploited by employing mainly the ‘room pillar’ method, to a smaller extent by ‘longwall mining’ or ‘room and pillar’ mining.

The common feature of all these types of deposits is the method of development and, in many cases, preparation of the deposit for exploitation.

Most often, the uranium deposits were developed through pits. In some cases, inclined shafts or tunnels were used. The system of horizontal development using cross tunnels and drift entries is to a great extent derived from the morphology of the ore body.

The last location where underground mining is still been done is GEAM Dolní Rožínka. In a mining area of 8.8 km² the last remaining mine, Rožná I, is being operated. This deposit is currently opened up to the 1200 m level (24th floor). Two mining methods have been employed at this deposit: overhand stoping and descending scaffolding to cave-in under an artificial roof. A total of 15.5 million tonnes of ore have been extracted here to date. The ore is processed at a chemical plant, the last of its type in the Czech Republic. The alkaline milling technology uses a classic procedure: grinding, leaching, sorption, elution and flocculation. The final product is ammonium diuranate, the quality of which meets international standards.

Following the termination of exploitation at individual deposits, the last step will be remediation of the consequences of mining operations, i.e. closeout of mine workings and restoration of the surface to its original condition and use.

4. HISTORY OF IN SITU LEACHING

Chemical in situ leaching (see Fig. 2) of uranium in the region of Hamr-Stráž has been done since 1967. During this period there have been many changes in the application of this method, brought about by the enormous development of the fields and boosted uranium production.

Unfortunately this method resulted in detrimental impacts on the environment, mainly the groundwater of the Cenomanian aquifer (Fig. 3). As a consequence, the water of the Turonian aquifer, being in the vicinity of high quality potable water sources, was also jeopardized. For this reason it has been decided to decommission and remediate the chemical plant.

5. DEVELOPMENT OF THE MILLING TECHNOLOGIES AT THE CHEMICAL PLANT

The original technique of chemical concentrate production from chemical leachates was derived from the process adopted for uranium ore milling plants. The basic principle of the process, i.e. sorption on ion exchangers, elution and ammonium diuranate precipitation, has not been changed. Throughout the

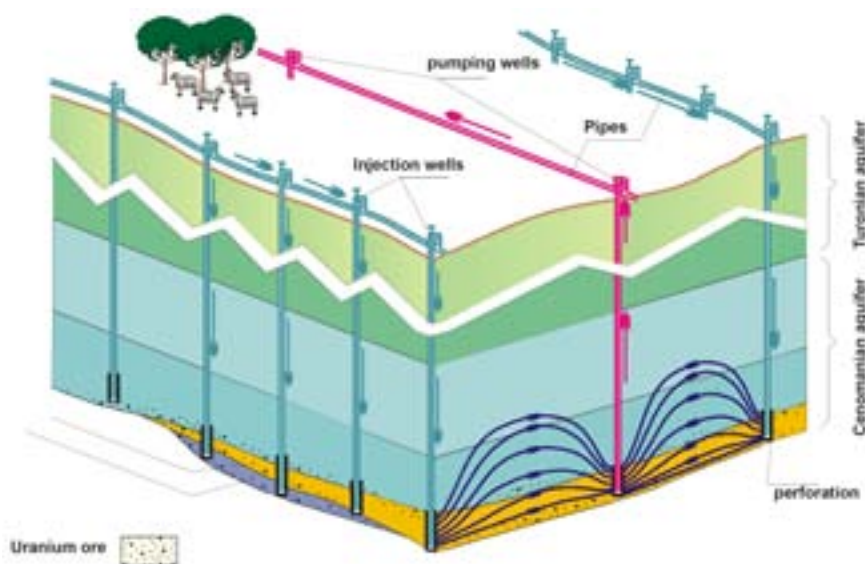


FIG. 2. Schematic of an in situ leaching field.

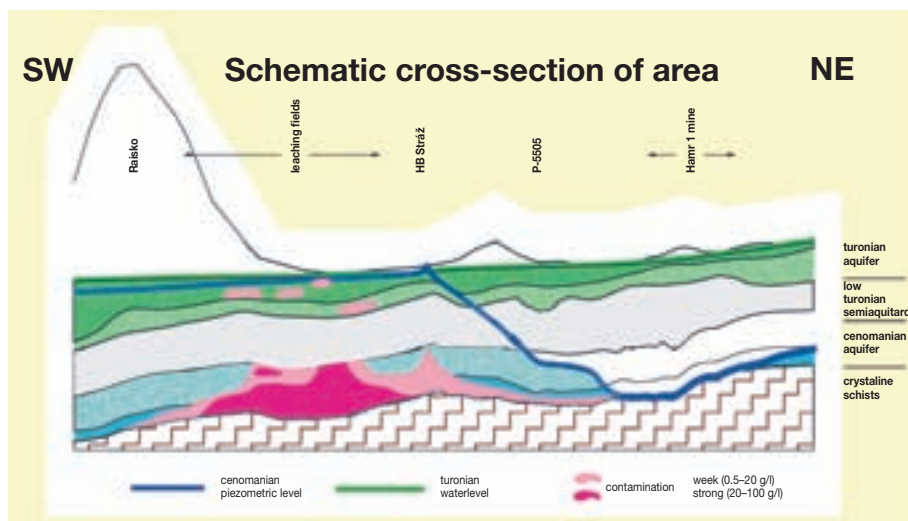


FIG. 3. Schematic cross-section of the deposit with the contamination plume.

operation of chemical leaching there has been technical development of individual installations as well as plant units (Fig. 4). The current high technological level and the level of automation makes it possible to produce uranium concentrate of standard quality, as required on world markets, at a lower cost than that at the start of production.

The first method employed made use of sorption columns with static beds, adapted from water treatment processes for water softening. The columns were treated against corrosion by rubberizing or were made of GRP. Extensive modifications were made to boost production. Semi-continuous columns were gradually set up. The filtration system separating the solution from ion exchange resin (originally sand beds) was changed. By removing part of the ion exchange resin it was possible to achieve contra-flow. This increased the efficiency of the columns significantly. The elution columns underwent similar development.

The original uranium concentrate of ammonium diuranate (ADU), so-called 'yellow cake', produced at the chemical plant contained the contractually agreed 40% uranium content. Developments in technologies leading to savings in chemicals resulted in a uranium content of more than 50%. It was decided that starting from 1 November 1990, ADU would be produced in a quality that met the parameters required for its sale on the world market. A standard of purity, ČSUP, was drafted, encompassing several standards produced by international companies processing ADU into fuel elements. The



FIG. 4. View of the chemical treatment plant and tailings ponds.

ADU production technology had to be adjusted accordingly to meet such stringent standards. Fractional precipitation of impurities was introduced and a uranium content in the ADU of up to 74% was achieved. The task for the remediation period is to maintain the quality of ADU even under changing operational conditions.

6. REMEDIATION OF THE STRÁŽ DEPOSIT

6.1. Overview

Remediation of the consequences of chemical leaching of uranium in Stráž pod Ralskem is a complex problem, consisting of a variety of partial solutions. The Czech government adopted two separate resolutions with respect to this matter, namely the Governmental Decree of 20 May 1992 No. 366, determining a transitory period between 1992 and 1994, including the characteristics of a special regime of exploitation, and the Government Decree of 6 March 1996, presenting the decision to terminate chemical leaching of uranium in Stráž pod Ralskem. Concurrently, the ministries and other state

agencies concerned approved the 'Framework Procedure for Decommissioning and Remediation of the Chemical Leaching and Programme of Verification Work at the Preparatory Stage of Remediation of the Locality and Rock Environment Affected by the Chemical Leaching of Uranium in the Region of Česká Lípa'.

Over the period of chemical leaching of uranium (approximately 32 years), a total of almost 5 million tonnes of sulphuric acid and other chemicals were injected. Most of the products (approximately 99.5%) of the reactions of the acids with the rocks are located in the Cenomanian aquifer, contaminating 186 million m³ of water in an area of approximately 24 km². Approximately 0.5% of the contamination is located in the Turonian aquifer. This has contaminated about 80 million m³ of water in an area of 7.5 km². There is no natural 'self-remediation' of contaminated groundwater. Presently, the groundwater in the Cenomanian aquifer is remediated by pumping, followed by evaporation at Station I (SLKR I) in order to prevent the spread of contaminants.

Assessment of the situation led the Czech government to the decision to terminate chemical leaching as of 1 April 1996 and to perform active remediation work, consisting of withdrawing contaminants from underground and their processing or environmentally safe disposal above ground.

6.2. Objectives of the chemical exploitation remediation

The objectives of the remedial activities are to:

- (a) Restore the rock environment to a condition guaranteeing continuing usability of Turonian water of the Northern Bohemia Cretaceous;
- (b) Decommission boreholes and surface installations;
- (c) Remediate the surface of the leaching fields to make them compatible with the regional ecosystem and the urban development plans.

6.3. Present status of the remediation of the Cenomanian aquifer

On the basis of the results of preliminary experiments, it has been decided to remove salts from contaminated solutions using an evaporation process, followed by crystallization of ammonia alum. Evaporation station I (SLKR I) is capable of processing almost 3 million m³ of contaminated solution and withdrawing more than 50% of the contaminant content at a production exceeding 200 000 tonnes of alum and about 350 000 m³ of stock liquor annually. The products of SLKR I will have to be further processed into

marketable products in a complex of evaporation station II (SLKR II) that is currently being prepared for gradual implementation.

A decisive step for the course of remediation work was the putting into operation of SLKR I in June 1996 (Fig. 5). Gradually, three evaporating columns were introduced, their guarantee and performance tests were conducted and the permanent operation of the entire complex was approved. Almost 3 million m³ of distillate were discharged into the Ploučnice River over the next four years. The concentrate from the evaporators was injected into the Cenomanian aquifer. Through the creation of a depression in the Cenomanian aquifer in the area of the leaching fields (Fig. 6) it was possible to stabilize the groundwater flow and to introduce overpressure of the Turonian aquifer in relation to the Cenomanian aquifer.

In 1996, the addition of sulphuric acid to injection solutions was terminated and the use of hydrofluoric acid was radically reduced. The so-called preparatory remediation period was completed by the commencement of the test operation of SLKR I, crystallization of alum, and putting into operation of SLKR II, with delivery of alum beginning in July 1999. By abstracting contaminants from the Stráž deposit it was possible to launch remediation of the chemical leaching of uranium. In 1999, a total of 2459 tonnes of alum were produced.



FIG. 5. View of SLKR I.



FIG. 6. View of the leaching field.

Decommissioning of the chemical leaching of uranium is a long lasting and complex process that must be continuously evaluated and specified. To this end, extensive monitoring, verification and modelling work has been carried out. The decommissioning of individual components is continuously being approved by the respective Czech authorities.

6.4. Further remediation projects

Remediation of the Cenomanian aquifer will be carried out employing the only feasible technological procedure, i.e. withdrawal of solutions and their desalination on the surface (Fig. 7). The major part of the treated water will be discharged into surface watercourses while another part may be injected back underground. Discharging into surface watercourses must meet the respective standards set down by State agencies. Discharging underground means discharging into mine water.

More than 80% of the contaminants are made up of components (SO_4^{2-} , Al, NH_4^+) of double ammonium aluminium sulphate–alum. Alum crystallizes from evaporator concentrates upon its cooling. It will be the main intermediate product from remediation and will be processed into economically usable products — aluminium oxide, pickle alum, ammonium sulphate and materials

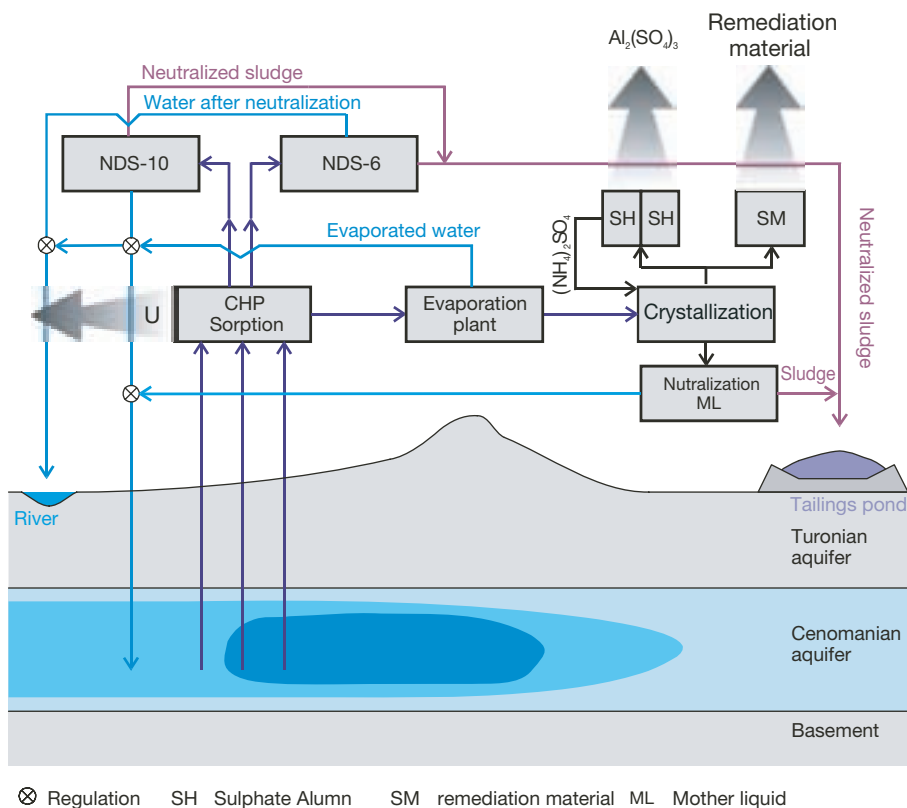


FIG. 7. Full layout of the remedial scheme.

for remediation of tailings ponds of chemical plants. This will bring about a radical reduction in the quantity of wastes to be disposed of in the region.

Because of the long time frame and technical and economic requirements of the remediation, the overall process is divided into five consecutive stages. This will make it possible to verify individual steps and direct the process in the subsequent stage so as to obtain the environmentally and economically best solutions.

The plant units for alum processing will have been constructed gradually prior to 2005 and will have a total capacity of about 200 000 tonnes of alum per year. The residue from crystallization (stock liquor) will be temporarily re-injected underground. The unit designed for final treatment (calcination and subsequent solidification) will be established by the year 2010. After that, no other substances will be re-injected underground. Pumping and treatment of solutions will continue until the target remediation parameters have been met.

The initial experience gained with the alum processing technology by DIAMO, s.p., indicates that it will be appropriate to consider a gradual construction of operating units with a maximum processing capacity of 100 000 tonnes of alum per year, provided that markets for the final products can be found.

7. SUMMARY AND CONCLUSIONS

The deposits in the area of Hamr-Stráž were discovered in the 1960s. Chemical leaching of uranium in the region of Hamr-Stráž began in 1967. This method resulted in detrimental impacts on the environment, mainly the groundwater of the Cenomanian aquifer. Over the period of chemical leaching of uranium (about 32 years), a total of nearly 5 million tonnes of sulphuric acid and other chemicals were injected underground. Most of the products (approximately 99.5%) of the reactions of the acids with the rocks are located in the Cenomanian aquifer, contaminating 186 million m³ of water in an approximately 24 km² area. Remediation of the Cenomanian aquifer will be carried out employing the only feasible technological procedure, i.e. withdrawing of solutions and their desalination on the surface. The expected duration of the remediation process will be about 30 years and the expected financial costs about 40 billion CZK (€1.25 billion).

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Paper 13

ENVIRONMENTAL EVALUATION AND REMEDIATION METHODOLOGIES OF ABANDONED RADIOACTIVE MINES IN PORTUGAL

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Abstract

The extensive information available from decades of exploration, exploitation and ore milling, as well as information acquired during characterization activities, have allowed the development of a database that includes data from 61 radioactive mining areas. Development of a management strategy for radioactive wastes is, generally, a complex process that aims at achieving a reasonable balance between two often conflicting objectives: maximization of risk reduction and minimization of costs. The process is one of optimization of the protection, in which the available alternatives are evaluated and compared, taking into account all the associated benefits and some restrictions, such as the setting of a maximum annual admissible radiation dose. It was necessary to define a methodology for the determination of the contaminated areas that allows the distinction between the anomalies in water, soil and stream sediments that are due either to natural processes or to mining activities. Multi-variant methods of analysis were used, capable of identifying and distinguishing the global geochemical signature inherent to natural processes from the anthropogenetic contamination. The results of that characterization made it possible in some cases to propose effective remediation projects, both in technical and economic terms, according to standards recommended by different international organizations.

1. EXPLOITATION OF RADIOACTIVE MINERALS IN PORTUGAL – CAUSES OF IMPACTS

From 1907 to 2001, five different entities were responsible for the exploitation of radioactive minerals in Portugal: Sociedade Urânio-Rádio (1912–1945), Companhia Portuguesa de Rádio, CPR (1945–1962), Junta de Energia Nuclear, JEN (1962–1977), Empresa Nacional de Urânio, EP, ENU (1977–1990), and Empresa Nacional de Urânio, SA (1990–2001). Until 2001, when production ceased, about 4370 tonnes of U_3O_8 had been produced (Fig. 1). In addition, radium salts were produced until 1945, and an estimated 13 million tonnes of different kinds of wastes were generated.

The exploited ore bodies had different characteristics, particularly in terms of typology and morphology (depth and regularity of the mineralization distribution), or host rock characteristics. These characteristics determined the different mining methods: underground, open pit, combined underground/open pit, sometimes with recourse to heap or in situ acid leaching. As a result of this activity, low grade ores and waste rock dumps, as well as tailings ponds, leaching wastes and sludges from effluent treatments, still exist in areas of uranium leaching [1].

The impact of waste dumps on soils, water, air, the landscape, as well as in terms of radiological impact, differs according to the nature of the exploited ores. The assessment of environmental impacts associated with the mining of radioactive ores requires additional investigations to those which deal with the environmental impacts associated with the exploitation of other metallic ores.

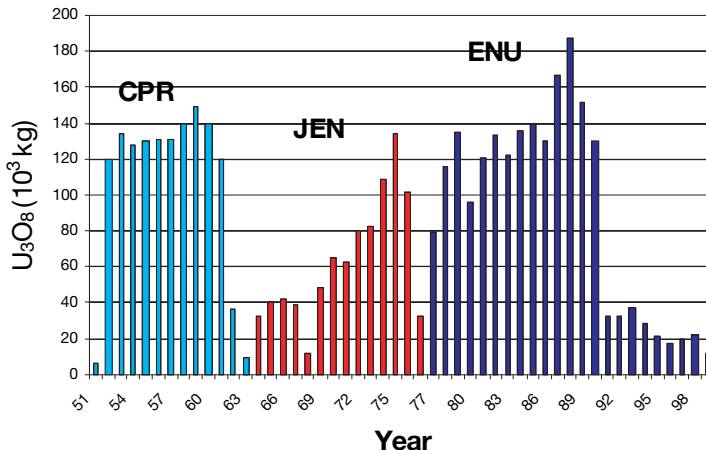


FIG. 1. Uranium concentrate production in Portugal.

Although mining and milling wastes contain only naturally occurring radionuclides, these cannot be considered to be in their original state or concentration. Exposures attributable to such wastes should not be regarded as exposure to natural background radiation [2].

Ore processing resulted in concentrates of about 80% U_3O_8 , while in the milling wastes, in addition to a small amount of uranium, significant amounts of other radionuclides were present. These wastes may also contain various amounts of other materials, such as those used in the milling operations or in the final treatment of wastes, as well as those associated with ore mineralogy. The first group includes sulphuric acid, sulphates, chlorides, carbonates, nitrates, ammonia, calcium hydroxide, manganese, organic solvents, while the one associated with the mineralogy quite often contains iron, copper, vanadium and arsenic, among others. Furthermore the treatment of waste water from mining and milling operations produces great amounts of $(Ba,Ra)SO_4$ sludge [3].

2. ENVIRONMENTAL REMEDIATION OF DEGRADED MINING AREAS – RESPONSIBILITIES

The seriousness of the situation and the urgency of finding appropriate means of re-establishing an environmental equilibrium in the areas affected by the mining activity led the Portuguese Government to assume, as its fundamental duty, the ensurance of adequate remediation of those areas of the continental territory of Portugal. This policy has been confirmed by the publication of Law Decree 198-A/2001, which establishes the main principles and objectives of the remediation and monitoring of degraded old mining areas. Said legislation established the public service nature of the corresponding remediation and granted the exclusivity of such activity to the EXMIN company — Companhia de Indústria e Serviços Mineiros e Ambientais, SA.

According to the concession contract the remediation of degraded and abandoned mines aims at an improvement in environmental, cultural, economic and regional conditions, guaranteeing the public interest and the preservation of environmental patrimony, through a set of interventions based on adequate levels of efficiency and quality and guided by entrepreneurial management criteria.

Following the ‘Despacho Conjunto’ 242/2002, dated 14 March 2002, recognizing the “undeniable public interest of an immediate state intervention on the area of uranium mining exploitation” an initial set of 30 uranium mines were considered under the scope of action of Law Decree 198-A/2001.

3. RADIOACTIVE WASTE MANAGEMENT STRATEGY

The development of a waste management strategy is usually a complex process that aims at achieving a reasonable balance between two often conflicting goals: maximization of risk reduction and minimization of financial expenditure. The process is based on the optimization of protection, for which available alternatives are evaluated and compared, taking into account all associated benefits and detriments as well as possible constraints such as an annual dose constraint.

According to the IAEA [4] the parameters that should be considered include:

- (a) Radiological and non-radiological impacts on human health and the environment before, during and after the remediation work;
- (b) Monitoring, maintenance and control before, during and after the remediation work;
- (c) Some restrictions on future use of property and water resources;
- (d) Costs associated with different alternatives and the resources available for implementing the alternatives;
- (e) Amounts of the various wastes to be managed;
- (f) Socioeconomic impacts, matters relating to public acceptance, including the degree of acceptance by the communities;
- (g) Good available engineering practices.

The different uranium mining areas were grouped in a multiparametric way according to:

- (1) Increase in local radiological levels (based essentially on external radiation values) as a result of moving radioactive materials from underground to the surface, presence of mill tailings, effluent treatment sludge, dumps of ores with different uranium grades according to the cut-off at the time of exploitation;
- (2) Existence of acid waters contaminated with radium and heavy metals in open pits and underground mines where acid leaching took place or in cases where sulphides are present in appreciable amounts;
- (3) Exploitation and processing magnitude (volumes, weight, spatial distribution of dumps and waste characteristics);
- (4) Situations presenting a risk such as unmarked access to underground works, geotechnical instability in certain waste dumps and tailings dams, as well as subsidence in underground mines;

- (5) Landscape impacts associated with waste dumps, open pits and tailings dams;
- (6) Other environmental factors (population centres, land water use).

With the work so far developed by EXMIN the following goals were met:

- (i) Significant improvements in effluent monitoring with a better level of control, guaranteeing more feasibility when evaluating the results of proposed remediation actions (post-monitoring remediation);
- (ii) Identification of the extension and degree of chemical and radiological contamination for those mines presenting a greater risk;
- (iii) Evaluation of geotechnical risks (slope stability and subsidence) in critical areas;
- (iv) Methodological definition of a strategic approach to the various environmental problems associated with radioactive mines;
- (v) Quantification and budgeting of complementary characterizations;
- (vi) Establishment of 'master plans' for some of the mining areas with preliminary evaluation of costs for interventions and post-remediation monitoring.

The alternatives evaluated considered the already regulated hydro-chemical and radiological parameters, while for those not presently regulated EXMIN proposes the adoption of criteria based on applicable international legislation.

Thus the proposed solutions took into account the following:

- (a) Radiological, hydrochemical and geochemical characteristics before exploitation;
- (b) Higher or lower degrees of restrictions on future use of land or water resources;
- (c) Costs of different remediation alternatives in a context of limited financial resources and in the 2000–2006 programming and application of the 3rd Portuguese Community Support Framework;
- (d) Increasing public sensitivity to radioactivity matters, making it difficult to obtain acceptance for transporting significant amounts of waste between different 'concelhos' (counties).

The lack of regional background radiological, hydrochemical and geochemical data in the uraniferous metallogenic Beiras Province required the selection of an area in the neighbourhood of Oliveira do Hospital and the proposal of a work plan to determine its background for use in comparisons

with the values obtained in the mining areas located in the same metallogenic province [3].

4. CHARACTERIZATION OF THE MINING AREAS – METHODS AND TECHNIQUES

4.1. Data collection

In order to manage the great amount of dispersed information resulting from decades of exploration, exploitation and ore processing, as well as information acquired during characterization activities, a database was established with all this information in a format that allowed its integration into a Geographical Information System [1]. The adopted methodology in building the geographical information system (GIS) for each mining area is illustrated in Fig. 2.

4.2. Sampling and radiological characterization

Water being one of the major radionuclide transport agents, its radiological monitoring in the neighbourhood of the mining areas was given special attention. Similarly, air monitoring for external radiation, radon and its progeny, as well as dust content, was also done.

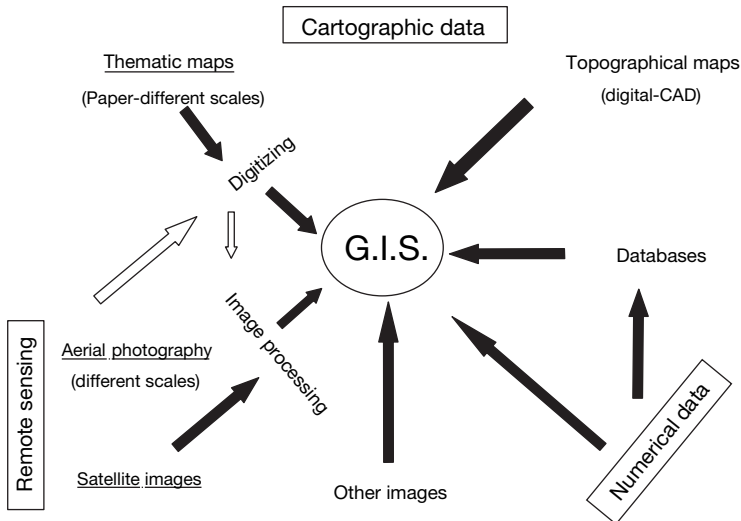


FIG. 2. Data integration in a GIS.

The main parameter used internationally to assess radiological risk is the effective dose that weighs the different organs and tissue sensitivities to given ionizing radiation.

In principle, the main interested international organizations consider that acceptable dose limits should be as low as reasonably achievable in the face of specific social and economic factors. The management of mining and milling wastes would require the implementation of measures that will provide acceptable protection of human health and the environment, within the requirements and recommendations of the IAEA and the ICRP, and governed by principles of justification, optimization and dose limitation.

Radioactive waste shall be managed in such a way that predicted impacts on the health of future generations will not be greater than the relevant levels of impact that are acceptable today [5].

It is required that for the critical group of members of the public an effective dose of 1 mSv/a attributed to practices is not exceeded, or under special circumstances an effective dose of up to 5 mSv in a single year, provided that the average dose over five consecutive years does not exceed 1 mSv/a [2].

Council Directive 96/29/Euratom establishes an effective dose limit of 1 mSv/a for members of the public allowing, however, in special circumstances that a higher effective dose may be authorized in a single year provided that the average over five consecutive years does not exceed 1 mSv/a. For exposed workers the limit for effective dose shall be 100 mSv over five consecutive years, subject to a maximum effective dose of 50 mSv in any single year. In the context of the implementation of the European Directive [6] the 'total added annual exposure' for a realistic exposed group was calculated. According to a current French regulation (Decree 90.222), compliance is achieved if the ATAER (added total annual exposure rate), based on a maximum annual exposure of 5 mSv, is less than 1. A proposal for detailed realistic calculation of the 'added annual effective' dose in mSv/a was presented to the Portuguese authorities, including the methodologies for the calculation of the dose values for the background reference area. 'Added dose' means that each exposure is due to the difference (Δ) between that of the area where the 'critical group' is located and the area representing natural background.

4.3. Methodologies for the definition of contaminated areas

The uraniferous ore deposits discussed in this paper are located in the Beiras metallogenic province where other deposits that are without economic potential are also frequently present and also contribute to the radiological background as a consequence of strictly natural causes. Both exploited and non-exploited ore bodies are of similar mineral paragenesis and hence are

frequently associated with sulphides, which are usually the cause of a high heavy metal background in the environment due to simple natural causes. Therefore, to define anthropogenetic contaminated areas, there was a need to first establish a methodology to allow them to be distinguished from other areas with anomalous background values due solely to natural processes.

Multi-variant analysis methods are needed to identify and distinguish the global geochemical signature inherent to natural processes from the anthropogenetic contamination. The methodology used was a discriminating analysis. The areas that show contamination induced by the old mine workings were clearly identified and mapped; moreover, groundwater contamination seems to be quite restricted geographically and depends on the main fractures. By the application of this methodology U, ²²⁶Ra, Cl, F, SO₄, Ca, Mn, Fe, as well as other less important elements such as Al, Be, As, Cs, Cu and Zn, were identified in many areas as the chemical signature of the mining activities.

5. METHODOLOGY FOR THE REMEDIATION OF ABANDONED URANIUM MINING RESIDUES

Several remediation techniques can be applied. However, given the quantities involved the more traditional methodologies that deal with the wastes on the surface, in situ containment or relocation to open pits followed by sealing are particularly adequate, since these methods isolate the wastes from the main dispersion agents — air and water — therefore ensuring their natural integration into the environment. Interventions can thus be systematized in terms of actuation in three main groups:

- (1) Removal of wastes to an engineered disposal facility;
- (2) In situ containment and sealing;
- (3) Removal to a local open pit (with or without sealing).

The necessary flooding of underground mines will require the development of adequate techniques and strategies. In addition, processing plants and sites will require decommissioning and dismantling.

6. PROPOSED PROCEDURE

Taking into account the different problems, the possible solutions and the nature and volume of the wastes, the proposed action procedure for radioactive mines is presented. The main aspects to be considered are:

- (a) All the mines are situated in the province of Beiras, distributed over 15 ‘concelhos’ (counties) in the districts of Coimbra, Guarda and Viseu;
- (b) All require remediation;
- (c) Their wastes present varied degrees of radiation and reactivity.

Consequently, it is justified to subdivide the mines into four geographical zones, each associated with mined-out pits, which might be used for relocating and isolating the wastes. In selecting the sites for waste management facilities EXMIN takes the following into account:

- (1) Natural background levels of radiation;
- (2) Climatology and meteorology;
- (3) Geography, geomorphology, demography and land use;
- (4) Structural geology and seismology;
- (5) Mineralogy and geochemistry;
- (6) Surface water and groundwater hydrology;
- (7) Public acceptance issues.

The Quinta do Bispo, Castelejo, Prado Velho and Mortórios zones were selected.

Tables 1–4 show, for each of these zones, the volume of the wastes to be relocated from existing mines, as well as the procedure to be used. The present 61 radioactive mining areas will be reduced to only 35 deposit areas that are duly confined.

7. CONCLUSIONS

A database was developed for 61 radioactive mining areas, using historical records and field data, as well as experimental results from samples analysed. From these data a hierarchy was established for the mining areas, according to the complexity of the environmental impacts.

Additional characterization work is being carried out for the more complex and higher risk cases, in order to improve the diagnosis of the nature and extent of the related environmental problems.

Tailings represent the greatest challenge, particularly in terms of long term management, owing to the large volumes produced and their content of very long lived radionuclides and heavy metals.

TABLE 1. PROPOSED PLAN OF ACTION FOR THE MINES OF THE QUINTA DO BISPO GROUP

Type	Mines	Mining Method	Volume of wastes (m ³)		External radiation (μGy/h)
			to deposit	in situ	
	Urgeirica - Escombreira da Barragem Velha	U	—	1 650 000	2.16
	Urgeirica - Depósito minérios	U	17 650	—	3.18
	Urgeirica - Escombreira de Sta. Bárbara	U	100 000	—	2.57
	Urgeirica - Escombreira da Barragem Nova	U	—	1 180 000	n.d.
	Cunha Baixa	U+OP	26 470	411 765	0.43 to 1.62
	Espinto	OP	30	147 060	0.36 to 1.15
	Vales	U	9 120	—	0.71 to 2.54
	Formiga	IMW-U	1 180	—	0.56
	Freixiosa	U	1 770	—	0.54
	Pinhal do Souto	U	28 355	—	0.50 to 1.20
	Vale Covo	U	590	—	0.66
	Valdante	OP	—	24 410	0.23 to 0.82
	Picoto	U+OP	—	24 120	0.36 to 0.99
	Vale da Abrutiga	OP	—	717 650	0.56 to 0.72
	Mondego Sul	OP	—	250 000	0.51
	Corga de Valbom	OP	—	30 060	0.33 to 0.64
	Póvoa de Cervães	OP	—	32 350	0.26
	Sevilha	OP	—	—	—
	Quita do Bispo	OP	58 825	823 530	0.68 to 2.05
	w/ acid effluents and partial removal of wastes to deposit	U	underground	n.d.	non determined
	w/ partial removal of wastes to deposit	OP	open pit	—	non applicable
	w/ total removal of wastes to deposit	IMW	incipient mining works		
	w/ total removal of wastes to local open pit				
	w/ wastes presenting non significative impacts				
	CONTAINMENT DEPOSIT				

TABLE 2. PROPOSED PLAN OF ACTION FOR THE MINES OF THE CASTELEJO GROUP

Type	Mines	Mining Method	Volume of wastes (m ³)		External radiation (μGy/h)
			to deposit	in situ	
	Castelejo	OP	—	696 480	0.42 to 0.82
	Barrôco I	OP			
	Picoto	OP			
	Vale da Abrutiga	OP			
	Mondego Sul	OP			
	Póvoa de Cervães	OP			
	Sevilha	OP			
	w/ acid effluents and partial removal of wastes to deposit				
	w/ total removal of wastes to local open pit		U	underground	n.d.
	w/ wastes presenting non significative impacts		OP	open pit	—
			IMW	incipient mining works	non determined non applicable

TABLE 3. PROPOSED PLAN OF ACTION FOR THE MINES OF THE PRADO VELHO GROUP

Type	Mines	Mining Method	Volume of wastes (m ³)		External radiation (μGy/h)
			to deposit	in situ	
	Bica	U	850	147 060	>1.14
	Senhora das Fontes	U - OP	n.d.	n.d.	0.58 to 2.60
	Rosmaneira	U	23 530	70 000	1.09 to 3.12
	Tentinolho	U	1 765	29 400	0.67 to 1.30
	Barracão	PP	7 300	—	1.64
	São Domingos	U	2 385	—	0.91 to 1.64
	Forte Velho	U	13 530	—	1.94 to 2.56
	Carrasca	U	3 700	—	0.67 to 1.30
	Vale d'Arca	U	23 530	—	0.77 to 2.16
	Alto da Várzea	U	12 060	—	1.14
	Coltos	U	1 800	—	0.93
	Barroca Funda	U	2 950	—	0.80
	Cruz da Faia	U	3 282	—	0.74 to 0.99
	Sentinela	U	30	—	1.16
	Pedreiros	U	4 120	—	0.55
	Ribeira do Ferro	OP	—	2350+?	0.24 to 1.44
	Barroco D. Frango	OP	—	59 400	0.33 to 0.68
	Pêra do Moço	OP	—	—	0.42
	Freixinho	OP	—	823 530	0.31
	Pai Moniz	IMW	—	—	—
	Tapada dos Mercados	OP	—	—	—
	Prado Velho	OP	59 420	232 350	0.34 to 0.86
	w/ acid effluents and partial removal of wastes to deposit				
	w/ partial removal of wastes to deposit				
	w/ total removal of wastes to deposit				
	w/ total removal of wastes to local open pit				
	w/ wastes presenting non significative impacts				
	CONTAINMENT DEPOSIT				
		U	underground	n.d.	non determined
		OP	open pit	—	non applicable
		IMW	incipient mining works	—	—

TABLE 4. PROPOSED PLAN OF ACTION FOR THE MINES OF THE MORTÓRIOS GROUP

Type	Mines	Mining Method	Volume of wastes (m ³)		External radiation (μGy/h)
			to deposit	in situ	
	Ervideira	EW	295	—	1.43
	Ferreiros	EW	2 120	—	1.12
	Fonte Velha	U	6 060	—	0.42 to 1.18
	Lenteiros	IMW-U	2 945	—	0.97
	Mestras	EW	2 945	—	0.92
	Reboleiro	IMW-U	9 120	—	0.26 to 0.96
	Carril	U	295	—	0.71 to 2.54
	Corgunha e Prazos	U	885	—	0.86
	Quinta das Seixas	U	600	—	0.70
	Alto do Cavalo	OP	—	117 650	0.74
	Maria Dónis	OP	—	69 470	0.33
	Alto da Raza	U	—	—	0.32 to 0.55
	Cótimos	U+OP	—	—	—
	Fontainhas e Gradiz	IMW	—	—	—
	Luz	U	—	65	—
	Mortórios	OP	29 415	114 705	0.40 to 0.81
	w/ total removal of wastes to deposit	U	underground	n.d.	non determined
	w/ total removal of wastes to local open pit	OP	open pit	—	non applicable
	w/ wastes presenting non significative impacts	IMW	incipient mining works	—	—
	CONTAINMENT DEPOSIT				

For some cases, it has already been possible to propose effective remediation solutions in both technical and economic terms, according to standards recommended by different international organizations. As engineering guidelines, maximization of the use of natural materials for containment and minimization of monitoring and maintenance needs after closure are to be adopted. To evaluate the long term efficiency of the planned scheme a numerical model was established and tested with the RESRAD [7] computer code.

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Paper 14

ABANDONED MINE SITE CHARACTERIZATION FOR REMEDIATION: THE CASE OF THE CUNHA BAIXA URANIUM MINE (VISEU, PORTUGAL)

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Abstract

Uranium mining activities at Cunha Baixa ceased in 1993 and a preliminary assessment of its chemical environmental impact was performed. Acid drainage affects surface and groundwater quality up to 1–1.5 km downward from the mining site. High levels of sulphate, Al, Mn, U, and low pH values (<4.5–5) make these waters unsuitable for irrigation and livestock watering. Irrigation of acid soils (pH <4.5) with contaminated waters presents risks to the crops owing to a high content of U in the available soil fraction. Consequently, maize harvested in these soils showed amounts of uranium in roots and leaves that may pose some risk when it is used for animal feeding and the plant residues are used for soil fertilization. According to the ‘tolerable daily intake’ of uranium, the low uranium content in corn allows it to be used to feed animals and for flour to make bread. *Thiobacillus ferrooxidans* were detected in mine water, but mine wastes submitted to static and kinetic laboratory tests (acid based accounting and a ‘humidity cell test’) did not show any capacity to generate acid drainage from sulphide oxidation throughout the testing period. Nevertheless, open pit mine wastes can be a source of water pollution in the Cunha Baixa mining area. Acid drainage can also be a residue from the heap leaching process used in the past to recover uranium from low grade ore.

1. INTRODUCTION

The history of radioactive ore mining in Portugal dates back to 1907 when prospecting and exploitation began for radium bearing ores. From 1944 to 2000, production centred on uranium. Mining activity developed in the Central-North regions of Portugal and produced about 4370 t U_3O_8 and 13 million t of waste rocks [1]. Nowadays, the environmental impact and remediation/rehabilitation of abandoned former uranium mining and milling sites is a major concern. The first step in the remediation process involves the characterization of mine sites in order to identify and evaluate aspects that have caused, or could result in, environmental damages.

The Cunha Baixa uranium mine (Viseu, Portugal), where mining activities ceased in 1993, was the abandoned mining site selected for study. The aim of this work was to evaluate the chemical and radiological water quality; to assess the potential risk to soils and plants from the use of contaminated water for irrigation; and to identify the sources of the problem that affects the water quality in the area surrounding the mine.

2. SITE DESCRIPTION

The mine is located approximately 20 km southeast of Viseu (Fig. 1), near Cunha Baixa village. The climate of the region is mild, with an annual average temperature of 13°C and rainfall of 1100–1400 mm. The agricultural land is currently used for growing maize, potatoes and greens.

Mining activities at Cunha Baixa started in 1970 and ceased in 1993. Mineralized veins and dispersed ore occur in an alkaline Hercynian granite and in metasedimentary schists from contact metamorphism. Autunite and torbernite are the main ore uranium minerals; metallic minerals such as pyrite, arsenopyrite and manganese oxides also occur in some veins.

The underground and open pit mining works (1970–1984) produced about 1000 t of U_3O_8 and one million tonnes of waste material were disposed of in a dump surrounding the mine area. The low grade ores produced in Cunha Baixa and adjacent mines (500 000 t) were placed in the open pit area and were subject to heap leaching (1984–1993) with sulphuric acid solutions (pH1–2). The uranium bearing liquors were pumped to the surface, where they were passed through adsorption resin columns. The loaded resins were transported to the Urgeiriça processing plant (25 km away from the Cunha Baixa mine) for elution.

The release of radioactive and toxic contaminants (^{222}Rn , ^{226}Ra , sulphate, metals) into the air and water can present hazards to residents and to the

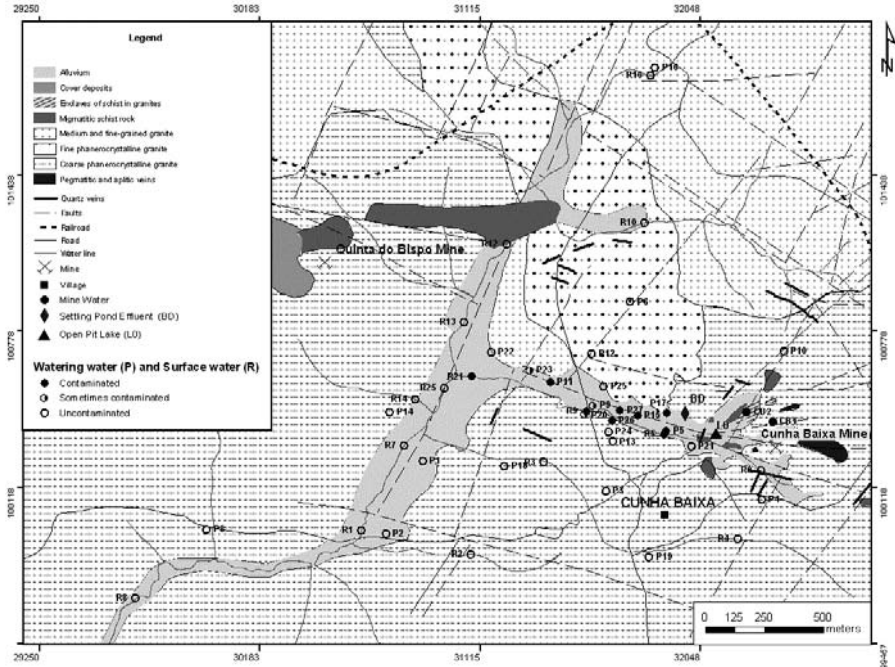


FIG. 1. Location of the Cunha Baixa study area and water quality characterization. Soils and plants studied are collected in the area irrigated by water wells P2, P5, P15, P17, P24 and P27.

surrounding environment, either during the active mining period or after mining activities have ceased. To minimize environmental damages related to mine workings, the Portuguese company Empresa Nacional de Urânio (ENU) took some actions, such as the revegetation of waste rock dumps with pine trees, and mine water treatment (applying a calcite/portlandite mixture to increase pH and adding a barium chloride solution to precipitate sulphate and radium). The neutralized water was discharged into a settling pond to remove solids and the effluents were released to either the waste rock dump area or into the open pit.

3. MATERIALS AND METHODS

Surface and groundwater were monitored seasonally (summer/winter) and sampled in private wells and mine shafts (Fig. 1) from 1995 to 1998. Water samples were filtered ($0.45 \mu\text{m}$) after in situ measurements of temperature, pH,

Eh, electrical conductivity (EC) and determinations of total alkalinity. The samples with or without HNO₃ acidification were stored under cool conditions at 4°C for further analysis.

Six composite samples from soils irrigated with private well waters (P2, P5, P15, P17, P24, P27) were collected from the surface to 30 cm depth in soil profiles located close to or further away from the mining area. Samples of maize plants growing in these soils were harvested 120 days after germination. The crops were not submitted to controlled conditions.

The physico-chemical characteristics of soils were determined in the fraction <2 mm after being dried at 40°C as follows: pH on a 1:2.5 soil/water mixture; total U by acid digestion [2]; and U-exchangeable by the ammonium acetate method at pH7 [3]. Maize plants were washed with distilled water, dried at 40°C, and ashed at 550–600°C. Analysis of the ashed plant material was done after digestion with HNO₃ and HCl. The results were expressed as plant ash or dry weight (DW).

Mine wastes (granite and schist rocks, sediments and drill core samples from the open pit area) were collected in the mining area to be submitted to static and kinetic tests according to Sobek et al. [4]. Static acid base accounting (ABA) and kinetic ‘humidity cell tests’ were carried out following the MEND procedures [5]. The ‘humidity cell test’ was performed over 6 or 12 cycles (42 or 84 days). Physical and chemical parameters (pH, Eh, EC, acidity, alkalinity, sulphate ions, and dissolved metals) were determined in the leachate recovered at the end of each cycle.

Chemical analyses for all samples were carried out by inductively coupled plasma mass spectrometry (cations), ion chromatography (anions), fluorimetry (uranium) and alpha counting after precipitation with barium sulphate (²²⁶Ra).

4. RESULTS AND DISCUSSION

4.1. Waters

Some of the chemical and radiological characteristics of the studied water samples are summarized in Table 1.

Mine waters (samples CB1 and CB2, cf. Fig. 1) classified as being of the calcium or magnesium sulphate type, were acidic (pH3–4) with high EC and ²²⁶Ra activity. High sulphate and heavy metal (Al, Mn, Fe, Zn, and U) concentrations were also found (Table 1). The acid mine drainage has the potential to contaminate waters in the area surrounding the mine.

Groundwaters sampled in the immediate surroundings of the mine (Fig. 1) had a calcium or magnesium sulphate type composition and are not

TABLE 1. CHEMICAL AND RADIOLOGICAL CHARACTERISTICS OF THE WATERS STUDIED IN THE CUNHA BAIXA MINING AREA

	pH	EC (μ S/cm)	SO ₄ ²⁻ (mg/L)	Al (mg/L)	Mn (mg/L)	Fe (μ g/L)	Zn (μ g/L)	U (μ g/L)	²²⁶ Ra (Bq/L)
Mine water	3-4	700-2800	300-2000	5-57	3-31	400-21 700	1020-2243	500-6000	1.70-8.60
Contaminated wells	3,6-5,1	1000-2700	575-1900	5-49	0,2-20	<5-120	429-3288	200-3500	0,45-5,97
Uncontaminated wells	5-7	35-558	3-211	0,02-0,13	0,01-0,12	<5	7-42	1-39	0,01-0,95
Contaminated surface water	3,7-5,9	745-2900	135-1892	0,74-16,39	0,33-6,76	<5-1 315	14-782	60-3200	0,40-2,00
Uncontaminated surface water	6,1- 7,2	82-925	14-291	0,01-0,18	0,01-1,54	<5-1 270	<5-105	2-21	0,06-0,47
Pit lake water	2,9-3,7	714-2880	355-2390	8,12-69,34	4,30-3450	960-36 640	509-2922	1200-6300	5,86-12,21
Settling pond effluent	7,8-8,8	1863-2920	1143-2017	0,02-1,16	1,26-5,88	<5-33	<5-23	222-2900	0,05-1,71

recommended for irrigation. The levels of some parameters (pH, EC, sulphate, Al, Mn, cf. Table 1) and total dissolved salts (640–2630 mg/L), fluorine (1–1.6 mg/L), Co (50–415 $\mu\text{g/L}$) were above the recommended or admissible maximum values established by Portuguese law for irrigation water [6]. The uranium content also exceeds the trigger values established, e.g. in Canada [7], and Australia and New Zealand [8] for irrigation (0.1 mg/L) and livestock drinking water (0.2 mg/L). The ^{226}Ra activity levels were normally lower than the trigger value (5 Bq/L) admitted in Australia and New Zealand [8] for irrigation water. The contaminated wells affected by acid drainage were located ESE-WNE along the groundwater flow direction, about 1 km away from the Cunha Baixa mine (cf. Fig. 1). The use of this water for irrigation presents risks to soils (acidity and salinity hazard) and to crops (phytotoxicity).

The pit lake water (Fig. 1) presented a chemical and radiological composition analogous to mine waters (Table 1). This acidic water (pH <3.5) percolating through permeable heap leaching wastes was able to dissolve minerals, mobilize elements and increase their dispersion. Thus the open pit area presents a risk to surface and groundwater by continuous contaminant leaching.

Surface water contamination was found only in the main drainage channel that drains the mine area approximately 1–1.5 km downstream (Fig. 1). Parameters such as pH, sulphate, Zn (Table 1) and Ni (18–176 $\mu\text{g/L}$) frequently exceeded the Portuguese regulatory limits for surface waters [6]. Uranium and ^{226}Ra activities in this surface water were higher than the other surface waters sampled (Table 1).

In spite of the efficient mine water treatment that removes the majority (50–80%) of the dissolved constituents, the discharging effluents can be a source of contamination due to high sulphate and U contents (Table 1), as well as fluorine (3–7.5 mg/L) and Ca (375–645 mg/L). The neutralization process was responsible for the hardness observed in surface and groundwater (total hardness >120 mg/L CaCO_3), a characteristic uncommon in alkaline–granitic areas such as the studied area.

4.2. Soils and plants

The use of contaminated water for irrigation can modify soil properties and increase its metal concentration, which may then accumulate to toxic levels. Vegetation grown in these contaminated soils can take up some of the metals, spreading contamination through the food chain [9].

The soils studied showed pH values ranging from 4 to 6.2, had low CEC (<6.38 cmol/kg), low clay (<160 g/kg) and organic matter content (<35 g/kg) [10], and a high content of total uranium (Table 2). Soil contamination cannot

be defined in relation to total metal concentration only, because even when the total content is similar in different soils, the species distribution of the metals is often quite different. Many authors have reported a good correlation between soil extractable metals and metal uptake by different test plants [11].

Among soils irrigated with contaminated waters, the more acidic soils near the mine area ($\text{pH} < 4.5$) showed an exchangeable uranium concentration that ranged between 35 and 70% of the total uranium soil concentration (Table 2). These soils present risks to the crops owing to the high available U concentration.

The metal concentrations in the plant tissues could reflect the concentration of soil in which the plants grow. However, this interrelationship is modified in many plants by tolerance mechanisms and other environmental factors. Mobility and phyto-availability of metals in soils depend on various soil characteristics such as pH, organic matter content and cation exchange capacity. The uptake also depends on the plant species and plant cultivars [12].

The roots of maize plants harvested in the mining area show higher U content (Table 3) than other plant tissues (leaf, stem or corn). This may be due to a defence strategy of the plants: immobilization of metals in the roots would be less dangerous for plant growth [13].

Levels of uranium in plant ash ranging between 0.5 and 2 mg/kg are considered to be normal levels for plants by many authors [14]. The U content in leaves of maize plants growing on soils irrigated with contaminated water exceeded the reported normal value (Table 3). The lowest uranium concentration was detected in maize harvested in soil irrigated with uncontaminated water and outside the mining area (Table 3). However, the uranium concentrations can also be considered as relatively high and may contribute to food chain contamination.

Maize harvested in soils irrigated with contaminated waters showed amounts of uranium in roots and leaves that may present some risk for animal feeding and for soils when the plant residues are used for soil fertilization, an agricultural practice common in this area.

According to the uranium 'tolerable daily intake' (TDI) of $0.6 \mu\text{g/kg}$ body weight per day proposed by the WHO [15], the low uranium content detected in corn ($< 26 \mu\text{g/kg DW}$; Table 3) allows it to be used to feed animals and in flours to make bread.

5. INVESTIGATIONS OF MINING WASTES

The ABA test was performed in order to determine the balance between acid production (AP) and acid consumption (NP) of the mining waste samples

TABLE 2. pH VALUES AND URANIUM CONCENTRATIONS IN SOILS OF THE CUNHA BAIXA STUDY AREA

Soil	pH (H ₂ O)	U _{total} (mg/kg)	U _{exchangeable} (mg/kg)
Acid soils irrigated with contaminated water	4.04–4.40	225–362	128–142
Soils irrigated with uncontaminated water	5.10–5.14	9.8–27	1.04–1.08

tested. To avoid overestimation of AP the parameter was corrected according to the procedures of Lawrence et al. [16]. Paste pH values also suggested that some samples had acidity before acid generation set in. The ABA test results and the values for the ‘neutralization potential ratio’ (NPR = NP/AP) showed that some samples (schist with pyrite and acid leaching open pit wastes) had the capacity to become acid generating materials [10]. These samples were submitted to the kinetic ‘humidity cell test’ which simulates geochemical weathering processes in order to evaluate drainage quality over the testing time. The mine wastes tested did not show a significant capacity to generate acid drainage from sulphide oxidation over the testing time. Nevertheless, *Thiobacillus ferrooxidans* was detected in the mine waters [10].

However, open pit wastes can be a source of water contamination in the Cunha Baixa mining area. The samples tested showed low acid neutralizing potential and a capacity to generate acid leachates (pH <4.5) with a potential to release sulphate and heavy metals (Al, Fe, Mn, Ni, Zn and U) [10]. Thus, acid drainage could still be the result of the heap leaching process used in the past to recover uranium from low grade ore.

6. CONCLUSIONS

A preliminary assessment of chemical environmental impacts resulting from the uranium mining activities in Cunha Baixa was performed. Acid drainage affects surface and groundwater quality 1–1.5 km downward from the mining site. The principal contaminants were sulphate, Al, Mn and U. These waters are not recommended for irrigation or livestock watering. Mine water treatment was responsible for water hardness. Open pit mine wastes can be a source of surface and groundwater contamination.

TABLE 3. URANIUM CONTENT IN MATURE MAIZE PLANT TISSUE HARVESTED IN THE CUNHA BAIXA SOILS STUDY AREA

Maize plants harvested	Root		Leaf		Stem		Kernels	
	($\mu\text{g/kg}$ plant ash)	(mg/kg DW^*)	($\mu\text{g/kg}$ plant ash)	(mg/kg DW)	($\mu\text{g/kg}$ plant ash)	(mg/kg DW)	($\mu\text{g/kg}$ plant ash)	(mg/kg DW)
In the mining area	209-950	32 612-59 320	1.8-19.2	689-1237	0.6-3	121-166	0.26-1	7-26
Outside the mining area	10	1124	1.2	43	0.16	7	(-)	

*DW not determined.

Acid soils (pH <4.5) irrigated with contaminated water introduced risks to crops owing to high U amounts in the available soil fraction. Maize harvested in these soils contained uranium concentrations in roots and leaves that may present some risk for animal feeding and for soils due to organic soil fertilization.

Acid heap leaching, used as a uranium recovery process, can also be responsible for the acid drainage that still occurs in the Cunha Baixa area, although mining activity ceased 10 years ago, and in spite of the remedial actions undertaken by ENU.

In the near future, other measures to minimize water percolation in the open pit area may be implemented, such as the coverage of heap leaching wastes with a neutralizing material (lime), followed by compaction and capping with a thick soil–clay layer.

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Paper 15

GLOBAL SENSITIVITY ANALYSIS — A TOOL FOR REMEDIATION

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Abstract

The deterministic model implemented in the Ecolego code for radioecological assessments of the near surface disposal site at Vaalputs has been used to perform a set of probabilistic calculations using the Monte Carlo method. The migration from the repository of the ^{14}C nuclide and of the nuclides of the ^{238}U chain has been analysed to assess the aqueous phase release scenario. Uncertainty and sensitivity analyses have been conducted and some of the results are presented in this paper. The first step of a ‘divide and conquer’ approach to the sensitivity analysis is illustrated. The conclusion is that only eight hydrogeological parameters have a considerable influence on the variance of the output regardless of time, so in the next step the Monte Carlo calculations of all other hydrogeological parameters will be locked at their best estimates. This will facilitate the analysis of future work, which is important because many parameters are also involved in the calculation of the final doses. Another conclusion of this work is that the capacity for Ecolego to undertake exploratory data analysis must be enhanced, enabling it to better tackle the large amount of output data resulting from Monte Carlo simulations.

1. INTRODUCTION

The vault safety case (VSC) of the IAEA’s ISAM project implements a conceptual model of radionuclide migration from a near surface radioactive waste disposal site, hypothetically located in the Vaalputs region in north-western South Africa [1]. In the context of conducting an intercomparison exercise of tools for performing radioecological risk assessments, the VSC calculation case was repeated for the aqueous phase release scenario using the Ecolego toolbox [2, 3]. In this work we examine the parameter driven

uncertainties by assigning probability distributions to a certain number of model parameters. We also undertake a global sensitivity analysis to rank the impact of the variable parameters on the model output. We address a single nuclide (^{14}C) and a decay chain (^{238}U), respectively. The calculations include a large number of compartments, parameters and nuclides. Given the complexity of this situation, a 'divide and conquer' strategy was required for the sensitivity analysis. Therefore, in the Monte Carlo calculations, we vary only the near field and far field parameters, keeping the biosphere parameters constant.

2. THE VAALPUTS REPOSITORY

This facility was designed for the disposal of low activity waste. It consists of two sets of ten concrete vaults with an approximate area of $170\text{ m} \times 210\text{ m}$. The waste is enclosed in 200 L drums. The voids in the drums are filled with grout and the drums are placed in concrete boxes, which are also backfilled with cement grout. The boxes are stacked in the vaults. A concrete lid will cover the repository, with the aim of limiting the infiltration of rainwater.

The geology of Vaalputs consists of Precambrian crystalline rocks covered by sediments. The near surface sediments are mainly red sand, aeolian sand, calcretised sand, gritty clay and red/greyish fluvial, gritty, sandy clay with gravel and quartz pebbles. Vaalputs is situated in a semi-arid region with plant and animal life characteristic of a semi-desert environment.

3. SYSTEM DESCRIPTION AND INPUT DATA

3.1. Overview

The system studied may be subdivided into a near field region (the engineered vaults), a geosphere consisting of an unsaturated and a saturated zone, and the biosphere. Water infiltrates the repository, coming into contact with the drums which, after a short period of corrosion, start leaching. The nuclides dissolved in the pore water of the cement leave the drums by advection and diffusion. The advection of contaminated water arising from infiltration is assumed to take place vertically downwards, carrying the contaminants to a zone unsaturated with water. From there they are transported by water to the saturated zone. It is assumed that water is extracted from a well in the saturated zone and that this water will be used for drinking and irrigation.

The facility is designed for an initial inventory of 1.0×10^{16} Bq. As pointed out previously, we address the migration of the nuclides of one decay chain

only: ^{238}U ^{234}U ^{230}Th ^{226}Ra ^{210}Pb ^{210}Po . The initial inventory of ^{14}C amounts to 1.0×10^{15} Bq and that of the decay chain to 5.0×10^{10} Bq of ^{238}U , and to 5.0×10^{10} Bq of ^{234}U .

3.2. Vault input data

Physical and chemical degradation causes a decrease in the sorption coefficients for the repository vault, which is assumed to be linear and stochastic with time. After 1000 years, the vault has completely degraded, whereupon it is assumed that the sorption coefficients remain low and constant (Table 1).

The Darcy flux q is taken to be zero up to 100 years, it assumes a value of 1.8×10^{-3} m/a between 100 and 500 years and increases to 1.8×10^{-2} m/a after that point in time. The porosity θ_w is equal to 0.2 (both for the waste and the concrete base) and the bulk density is 1600 kg/m^3 .

Table 2 gives the parameters of the unsaturated zone, with the exception of the twenty-four variable sorption coefficients in the different regions of the unsaturated zone. These sorption coefficients are described by uniform distributions, using the best estimate values [3] to create an interval for the probability density functions (PDFs). The maximum and minimum values of the PDFs are equal to one and a half times and half of the best estimate for the PDF, respectively. The non-nuclide specific parameters of the saturated zone that are kept constant are: the hydraulic gradient (0.1), the bulk density (2000 kg/m^3), the dispersion length (30 m), the irrigated area ($1.0 \times 10^5 \text{ m}^2$), the depth of irrigation (0.5 m) and the volume of water extracted for other

TABLE 1. DISTRIBUTION COEFFICIENTS (K_d) ASSIGNED TO THE VAULTS

Element	$K_d \text{ (m}^3\text{/kg)}$	
	Non-degraded	Degraded
C	2	0.2
U	2.0	0.1
Th	5.0	1.0
Ra	0.05	0.05
Pb	0.5	0.05
Po	0	0

TABLE 2. PARAMETERS FOR THE UNSATURATED ZONE

Material	Depth (m)	Bulk density (kg/m ³)	Porosity (-)	Porosity distribution			
				Type	Minimum	Mode	Maximum
Red sand/ calcrete	2.7	1989	0.2	Triangular	0.16	0.20	0.24
Brown sand gritty clay	8.5	2230	0.2	Triangular	0.16	0.20	0.24
White kaolinite clay	8	2160	0.2	Triangular	0.16	0.20	0.24
Weathered granite	35.8	1683	0.2	Triangular	0.16	0.20	0.24

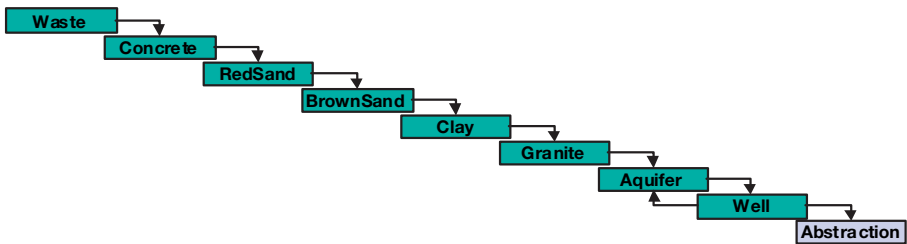


FIG. 1. The matrix interaction showing the transfer of mass between the compartments.

purposes (3300 m³). We assume that the hydraulic conductivity follows a uniform distribution (range 900–2700 m/a) and that the porosity can be described by a triangular distribution with a minimum equal to 0.20, a mode of 0.25 and a maximum of 0.30. The distribution coefficients (m³/kg) of the weathered granite in the saturated zone have a uniform distribution and are in the range of: 2.5×10^{-3} – 7.5×10^{-3} (C), 1.5×10^{-3} – 4.5×10^{-3} (U), 1.5–4.5 (Th), 0.25–0.75 (Ra), 0.15–0.45 (Pb) and 0.75×10^{-1} – 2.25×10^{-1} (Po).

4. THE CONCEPTUAL MODEL AND ITS IMPLEMENTATION IN ECOLEGO

We use the interaction matrix technique (MI) [4] to conceptually describe the migration processes between the near field, far field and biosphere. The interaction matrix is shown (in a compact form) in Fig. 1.

This MI is implemented in Ecolego through a graphics interface. Ecolego uses that information to automatically generate the code of the system of ordinary differential equations that describe the transfer processes between compartments with the help of transfer rates. For example, the inventory of the compartment changes with time at a rate given by Eq. 1:

$$\frac{dN_i}{dt} = \left(\sum_{j \neq i} \lambda_{ji} N_j + \lambda_N M_i + S_i(t) \right) - \left(\sum_{j \neq i} \lambda_{ij} N_i + \lambda_N N_i \right) \quad (1)$$

where i and j are compartments, and N and M are the amounts (Bq) of radio-nuclides N and M in a compartment. M is the precursor of N in a decay chain. The time dependent external source of N (Bq/a) is represented by $S(t)$. λ_N is the decay constant (a^{-1}) of nuclide N and λ_{ji} and λ_{ij} are the transfer coefficients (a^{-1}) representing the gain and loss of nuclide N from compartments i and j , respectively. By solving the system of equations given above, we obtain the time dependent inventory of each compartment.

5. RESULTS AND DISCUSSION

5.1. Uncertainty analysis

We used the simple random sampling option of Ecolego to conduct the uncertainty analysis. One thousand Monte Carlo realizations were performed. The input distributions are propagated through the chain of compartments, resulting in probability distributions. As an illustration, the histograms of the flux of ^{14}C from the vault (before entering the unsaturated zone) and from the unsaturated zone at/after 50 000 years, are shown in Fig. 2. It can be observed how the uncertainty — expressed in terms of the standard deviation of the flux distribution — increases from the repository to the unsaturated zone.

Figure 3 shows the time independent distribution of the nuclides of the ^{238}U decay chain. The variance in the results expressing the uncertainty in the spread of the input parameters is, in fact, only moderate if compared with the case of deep repositories.

The mean well water concentration as a function of time is shown in Fig. 4. The left panel of that figure shows the mean value and the 5 and 95 percentile curves of the concentration of the ^{14}C nuclide. The right panel shows the mean concentration of ^{14}C in the well water as a function of time, together with the corresponding concentrations for the ^{238}U decay chain.

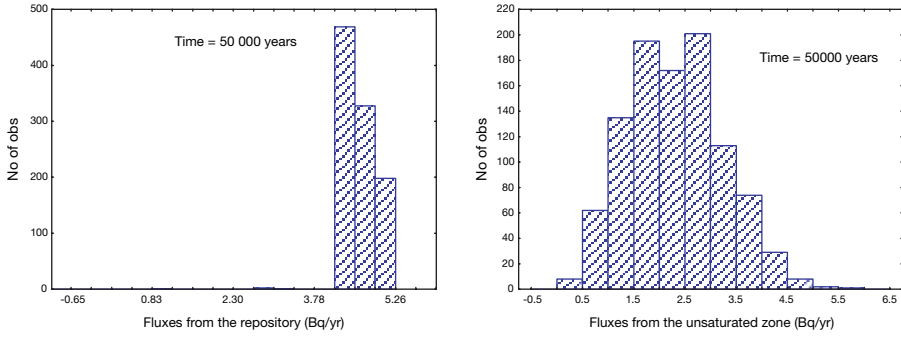


FIG. 2. Fluxes from the nuclides leaving the repository (to the left) and leaving the unsaturated zone (to the right). Observe the use of a logarithmic scale.

5.2. Sensitivity analysis

For the purpose of this paper we considered the water concentration in the well of the saturated zone as the endpoint of the ISAM liquid release scenario. The strategy adopted in conducting the calculations presented here was to ‘divide and conquer’. We studied the influence of the repository and hydrogeological parameters, to rank them according to their impact on the output. In a second step (which has yet to be performed), it will be possible to disregard the uncertainty of those hydrogeological parameters that have very little influence on the concentration. These parameters will be kept constant and new Monte Carlo calculations will be performed, now varying the main biosphere parameters together with the few hydrogeological parameters that are shown to have a considerable impact on the concentration in the well.

Using Spearman rank correlation coefficients it was possible to order the parameters that have a clear correlation with the output for the concentration of ^{14}C in the well water regardless of time. At a confidence level of 95% the ranking of the hydrogeological parameters in decreasing order of importance is (Table 3): $K_{d(\text{granite})} (^{14}\text{C}) > K_{d(\text{brown band})} (^{14}\text{C}) > K_{d(\text{clay})} (^{14}\text{C})$. The correlation between the flux of the unsaturated zone and the porosity θ_w was statistically significant, but the impact of this parameter on the water concentration was negligible.

Figure 5 illustrates the correlation between the ^{14}C concentration in the well water and the sorption coefficient of granite. The negative signs of the correlation values show that the higher the sorption coefficients, the lower the concentration, and thus the lower the impact.

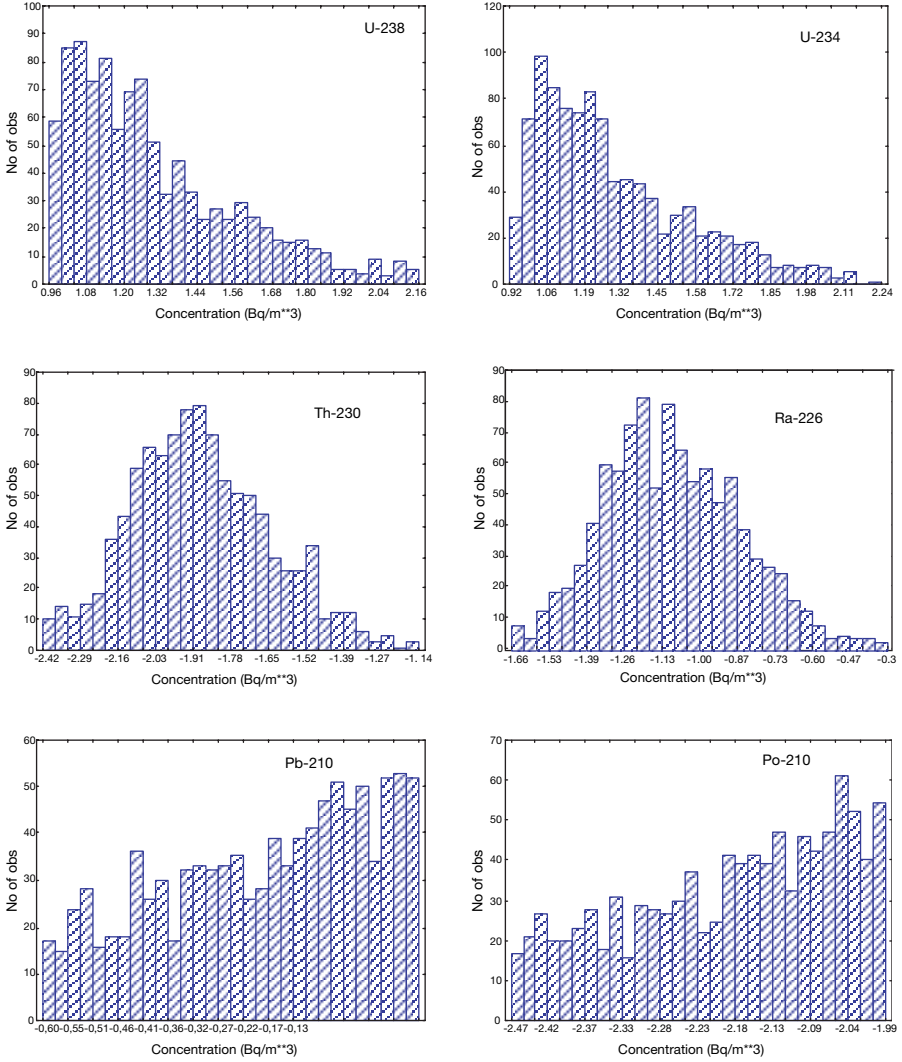


FIG. 3. Histograms of the time independent concentration of the ^{238}U decay chain.

A similar sensitivity analysis was carried out for the nuclides of the ^{238}U decay chain. Table 4 shows the results for the concentrations in the well water for these nuclides regardless of time. It is concluded that the parameters with a higher impact on the concentrations are, in decreasing order: $K_{\text{d}(\text{clay})} (^{230}\text{Th}) = K_{\text{d}(\text{saturated})} (^{210}\text{Po}) > K_{\text{d}(\text{saturated})} (^{210}\text{Pb}) > K_{\text{d}(\text{clay})} (^{226}\text{Ra}) > K_{\text{d}(\text{saturated})} (^{226}\text{Ra})$.

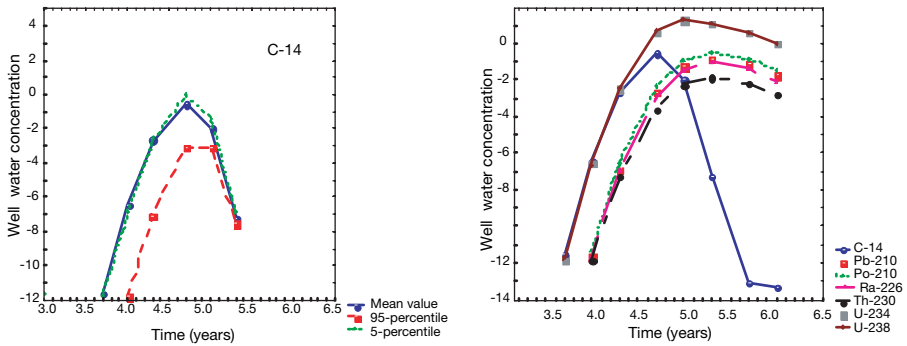


FIG. 4. Mean peak of the well water concentration of ¹⁴C and members of the ²³⁸U decay chain vs. time. The left hand graph also shows the 5 and 95 percentiles of the ¹⁴C concentration.

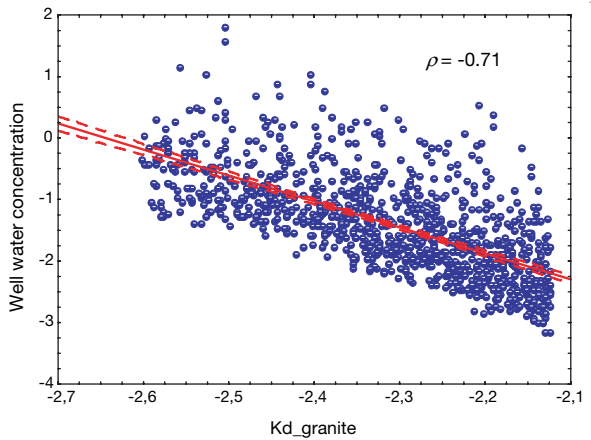


FIG. 5. Scatter plot of the correlation between the K_d coefficient of ¹⁴C for granite as a function of the concentration in well water.

TABLE 3. SPEARMAN CORRELATION COEFFICIENTS BETWEEN PARAMETERS AND INTERMEDIATE FLUXES OR THE CONCENTRATION IN THE WELL REGARDLESS OF TIME

Parameter	θ_w	$K_{d(\text{clay})}$	$K_{d(\text{Brown sand})}$	$K_{d(\text{granite})}$
Flux from the vault		-0.07		
Flux from the unsaturated zone	-0.07	-0.12	-0.21	-0.70
Well water concentration		-0.12	-0.21	-0.71

TABLE 4. SPEARMAN CORRELATION COEFFICIENTS BETWEEN PARAMETERS AND WELL CONCENTRATION

Nuclide	$K_{d(\text{saturated})}$	$K_{d(\text{brown sand})}$	$K_{d(\text{clay})}$
^{238}U	—	—	—
^{234}U	—	—	—
^{230}Th	—	—	-0.46
^{226}Ra	-0.18	—	-0.30
^{210}Pb	-0.42	—	—
^{210}Po	-0.46	-0.07	—

Although statistically significant, the correlation between the sorption coefficient of the brown sand ($K_{d(\text{brown sand})}$) and the concentration regardless of time of ^{210}Po in the well is too low, and therefore this parameter is not an important one.

The results presented here are, however, not complete. In fact, the impact of the parameters on the output often varies with time [5, 6] and, therefore, it will be necessary to calculate these impacts for different time points before excluding any parameter from the second step of the sensitivity analysis.

6. SUMMARY AND CONCLUSIONS

Ecolego was used to perform Monte Carlo calculations with the intention of performing an uncertainty and sensitivity analysis of the IAEA Vaalputs model for the liquid release scenario. The large amount of input data required the partitioning of the analysis (through the adoption of a ‘divide and conquer’ approach) into two steps. In the first step presented in this paper it was concluded that only eight hydrogeological parameters had a considerable impact on the variance of the nuclide concentration (regardless of time) in the well, which was used to abstract drinking and irrigation water.

However, the variation with time of the impact on the output of all variable parameters will require an extension of the present work. In fact, the many parameters, compartments, nuclides and decay chains imply large input data sets and complicate not only the uncertainty but also the sensitivity analyses. Also, the Monte Carlo simulations generate a huge amount of information. Therefore, the exploratory data analysis capability of Ecolego must be enhanced. This may be done by the inclusion in Ecolego of an exploratory data

analysis tool that calculates and presents graphically and by default many of the results needed for a first overview after the performance of the Monte Carlo simulations. In this way it will be possible to minimize the need for a time consuming interactive uncertainty and sensitivity analysis.

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Paper 16

ENVIRONMENTAL HAZARDS AT THE FORMER URANIUM MINES URGEIRIÇA, CUNHA BAIXA AND QUINTA DO BISPO

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Abstract

The exploitation of radium salts and uranium in Portugal over almost a century produced a large amount of waste that may be the source of environmental impacts in the surrounding areas. The mines of Urgeiriça, Cunha Baixa and Quinta do Bispo were the most important, with a total of 7.2 million t of mining mill tailings and wastes produced and currently deposited in several places near the old mining sites. A total of 325 samples from the tailings, waste waters, surface and groundwaters, stream sediments and soils were collected and analysed for total U and for the radionuclides of its decay chain. The comparative study revealed differences between the Urgeiriça, Cunha Baixa and Quinta do Bispo mining areas concerning the distribution pattern of the radionuclides. The most relevant difference is the high concentration of ^{226}Ra measured in waters and soils around the Cunha Baixa site, the result of geological and anthropogenic factors. The radionuclide activities in the stream sediments of the main watercourse that drains the Urgeiriça mine site are also high, particularly in the cases of ^{238}U and ^{230}Th , probably originating from the mill tailings deposit.

1. INTRODUCTION

Exploitation of radioactive ores in Portugal began in 1913, initially with the extraction of radium salts and after 1950 focusing on the extraction of uranium. It ended in 2000. Mining operations took place at 61 different locations in central Portugal and a large amount of waste, estimated at about 13 million t [1], was produced and deposited near the old mining sites.

The water that percolates through the old mines and the tailings, mechanical erosion, as well as radon exhalation from tailings, are the main potential sources of contamination of the environment. These processes can transfer chemical elements as well as radioisotopes to the surrounding environmental compartments.

The goal of the present work is to compare the degree and extension of radionuclide contamination in the vicinity of the former Urgeiriça, Cunha Baixa and Quinta do Bispo uranium mines. These were the most important exploitations in Portugal, carried out in Urgeiriça by underground work and in situ acid leaching; in Cunha Baixa both as underground and open pit mining; and in Quinta do Bispo exclusively as open pit [1]. Sulphuric acid heap leaching was used in the latter two mines for uranium recovery. These mining areas account for a total of 7.2 million t of the wastes that are currently deposited in several tailings. The three mines are also representative, for the most part, of the geological setting of the uranium metallogenic province of central Portugal. Uranium from the richest uranium ores (>500 ppm) of Urgeiriça, Cunha Baixa, Quinta do Bispo and other uranium mines was recovered, by dynamic acid leaching, at a single location near the Urgeiriça mine.

2. GEOLOGICAL SETTING

Mineralization in Urgeiriça occurs as siliceous-iron-pyrite-galena-pitchblende-type veins, intrude into a NE-SW fault that cuts a porphyritic medium to coarse grained biotite granite (Fig. 1 (a)). In Cunha Baixa, the mineralization is predominantly composed of hexavalent uranium minerals and occurs in quartz veins and tectonic breccias with N 40° E and N 70° W trends; these veins also cut a porphyritic medium to coarse grained biotite granite (Fig. 1 (b)). A mineralized metasedimentary enclave or roof pendant occurs immediately over the vein type mineralization and was exploited as an open pit mine. In Quinta do Bispo, the mineralization, similar to that of Cunha Baixa, occurs disseminated in a metasedimentary enclave in the same type of granite (Fig. 1 (b)).

Other types of rock, such as porphyritic or non-porphyritic granites, occur in both areas, as well as small veins of aplites, pegmatites and granite porphyry, and tertiary sedimentary deposits. Detailed geological information is provided in Refs [1–3].

All the mine sites are located in small catchment areas, namely the Ribeira da Pantanha in the case of the Urgeiriça mine, and Ribeira do Castelo for Cunha Baixa and Quinta do Bispo. Both are tributaries of the river Mondego, the main stream in this area of central Portugal.

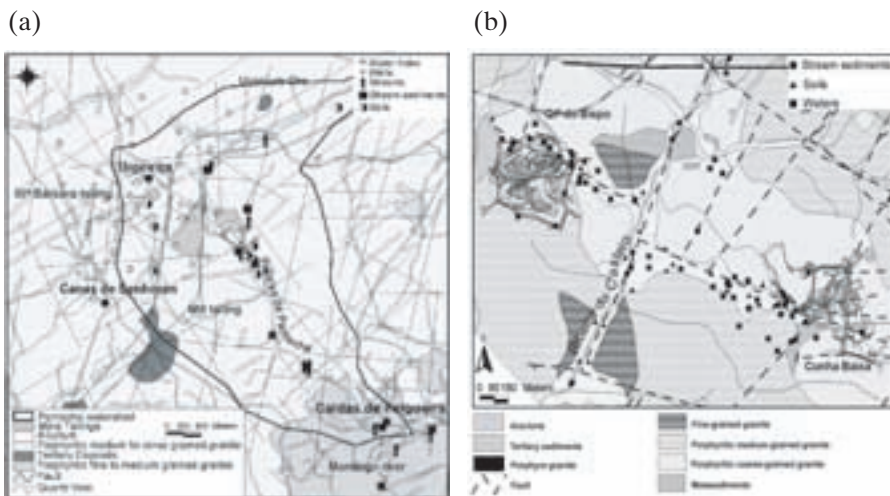


FIG. 1. Geology of the Urgeiriça (a), Cunha Baixa and Qrª do Bispo (b) mines and location of the samples collected to evaluate contamination by radionuclides.

3. MATERIALS AND METHODS

The sampled set comprises stream sediments (17), soils (48) and surface and groundwaters (216). The stream sediments were taken in the dry season and soil samples from different layers: one from the top 15 cm and the other from a depth between 15 and 30 cm. The water samples were taken from streams (26), wells (118) and monitoring wells (72), in two different campaigns (June and November 2000). The sources of contamination were also sampled as follows: geological materials from the mill tailings (32), wastewater of the mine and seepage from the tailings (8), and effluents from the treatment plant discharged in the two tributaries of the river Mondego (4). A detailed description of the analytical techniques is given in Ref. [2].

4. RESULTS

4.1. Contaminant sources

4.1.1. Tailings

The available data for U and ^{226}Ra in samples from several tailings located near the Urgeiriça, Cunha Baixa, and Quinta do Bispo mines are listed in Table 1.

TABLE 1. U AND ^{226}Ra IN TAILINGS FROM VARIOUS MINES (DATA OF THE FIRST TWO MINES TAKEN FROM REF. [4])

	Mine	Number of samples	U (ppm)	^{226}Ra (Bq/kg)
Tailings (low grade ore)	Cunha Baixa	13	318	1 950
	Quinta do Bispo	4	325	—
Mill tailings	Urgeiriça (unit A)	4	104	4 898
	Urgeiriça (unit B)	21	223	37 023
	Urgeiriça (unit C)	3	1523	3 360

The highest average uranium values occur in the non-outcropping unit C that is part of the Urgeiriça mill tailings deposit. As described in previous work [2], this unit is of small size and composed of precipitates from an old settling basin. The other units of the same tailings consist of sludge from the treatment plant, where uranium was selectively extracted from the ore and, for that reason the U content is the lowest of the whole set. On the contrary, the materials from the tailings of Cunha Baixa and Quinta do Bispo were not submitted to this kind of treatment, which explains the higher U content observed, with average values typical of low grade ore.

The mill tailings that have the highest ^{226}Ra values, being particularly enriched in this radionuclide in the fine grained unit B, are composed of clay and silt materials [2], meaning that ^{226}Ra was not completely removed from the sludge by the technological treatment procedures.

4.2. Contaminated waters

The water that percolates the interior of the underground mines, as well as the tailings located in the vicinity, can leach significant amounts of contaminants and transfer them to the surrounding environment. To minimize this risk a hydraulic system was built to divert the wastewater to treatment plants, with the aim of pH control and removal of ^{226}Ra , as well as other metals, before the release to the natural system [1–3]. Table 2 and Fig. 2 show the analytical data available for total U and ^{226}Ra , measured in samples of water from the underground mines, tailings seepage, as well as effluents from the treatment plants.

TABLE 2. ANALYTICAL RESULTS FROM MINE WATER (WM) COLLECTED IN VARIOUS MINES, FROM SEEPAGE FROM THE TAILINGS (WE), AS WELL AS TREATED EFFLUENTS DISCHARGED TO THE PANTANHA AND RIBEIRA DO CASTELO WATERCOURSES (WL)

		U			²²⁶ Ra		
Campaign		Urgeiriça	Cunha Baixa	Quinta do Bispo	Urgeiriça	Cunha Baixa	Quinta do Bispo
WM	June	140	1855	—	4.45	11.10	—
	November	117	597	—	5.58	10.33	—
WE	June	2340	1263	645	0.16	6.41	0.26
	November	1560	271	301	0.16	7.50	0.28
WL	June	0.8	1478	—	0.96	0.10	—
	November	17.0	186	—	0.26	0.53	—

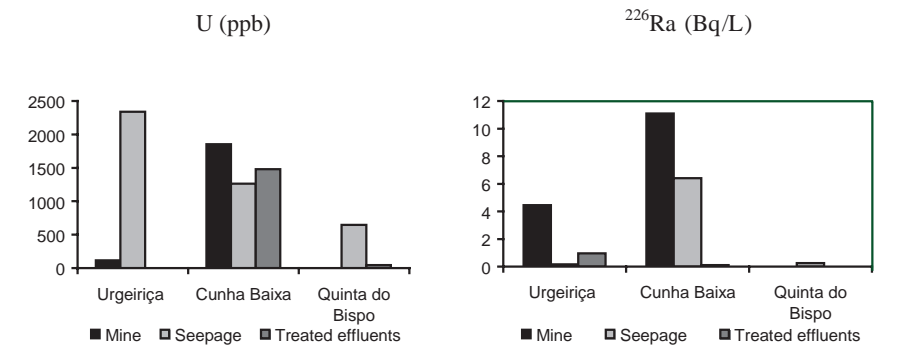


FIG. 2. U and ²²⁶Ra averages in wastewaters and treated waters measured in June 2000 at the Urgeiriça, Cunha Baixa, and Quinta do Bispo sites.

The drainage water from the Cunha Baixa underground mine carries higher concentrations of radioactive elements compared to the water that percolates the Urgeiriça mine. The U concentration factor varies between 5 and 13, while for ²²⁶Ra it is close to 2.

The seepage found in the various tailings also shows high U concentrations, with the highest values obtained in the water collected from the Urgeiriça mill tailings and the lowest in Quinta do Bispo. The highest ²²⁶Ra activity was measured in waters that discharge from the Cunha Baixa tailings, and the

lowest from the Urgeiriça mill tailings. The geological materials of this last tailings contain the highest ^{226}Ra activity, which suggests that only a small fraction of the radium in the solid phase transfers to the groundwater.

Based on the available data, it can be concluded that the procedures in the wastewater treatment plants are effective in the removal of ^{226}Ra , with more than 80% of the radionuclide removed from the wastewater. In the Urgeiriça plant uranium is also retrieved from the contaminated water in a proportion close to that obtained for ^{226}Ra . However, the control measures in Cunha Baixa do not seem to be as effective with respect to the removal of uranium, as more than 70% of the radioelement remains dissolved in the treated water and is discharged to the natural system.

4.3. The radionuclides in the surrounding environment

4.3.1. Surface and groundwater

The results obtained from water samples collected near the mine sites are shown in Table 3 and Fig. 3. In general, the radioactive elements have higher concentrations in boreholes and lower ones in surface waters. In the Cunha Baixa–Quinta do Bispo area the U concentration is also high in wells. A strong variability of concentrations was detected, as shown by the standard deviation values.

A comparison between the data from both mining areas reveals that the groundwaters of Urgeiriça contain less U and particularly ^{226}Ra than the groundwaters that circulate around the Cunha Baixa and Quinta do Bispo mines (Fig. 3).

4.3.2. Stream sediments and soils

The activities of U chain radionuclides in these environmental compartments are relatively high and show significant differences between the various mining areas, as Table 4 illustrates.

The stream sediments from the Urgeiriça area and soils from the Cunha Baixa and Quinta do Bispo area have higher average values of radionuclide activities (Fig. 4). The isotopic disequilibrium is remarkable in both environmental compartments. The ^{230}Th enrichment observed mainly in the stream sediments of Urgeiriça is noticeable; ^{226}Ra and ^{210}Pb show higher activities in soils of the Cunha Baixa and Quinta do Bispo area, but in these samples the ^{238}U activity is slightly lower than that measured in Urgeiriça.

TABLE 3. MEAN AND STANDARD DEVIATION FOR RADIOACTIVE ELEMENTS MEASURED IN WATERS COLLECTED AROUND THE FORMER URGEIRIÇA AND CUNHA BAIXA–QUINTA DO BISPO MINES

Campaign		U (ppb)		^{226}Ra (Bq/L)	
		Urgeiriça	Cunha Baixa– Quinta do Bispo	Urgeiriça	Cunha Baixa– Quinta do Bispo
Streams	June	8.5 ± 14.4	5.7 ± 6.8	0.14 ± 0.18	0.08 ± 0.11
	November	19.9 ± 30.1	1.8 ± 1.3	0.09 ± 0.12	0.02 ± 0.02
Wells	June	13.1 ± 40.7	84.6 ± 214.1	0.14 ± 0.23	0.57 ± 0.61
	November	16.5 ± 48.8	72.8 ± 114.2	0.13 ± 0.17	1.31 ± 1.77
Holes	June	38.4 ± 71.3	46.1 ± 113.8	0.20 ± 0.22	1.62 ± 3.87
	November	35.5 ± 82.6	22.0 ± 51.0	0.20 ± 0.22	2.01 ± 5.01

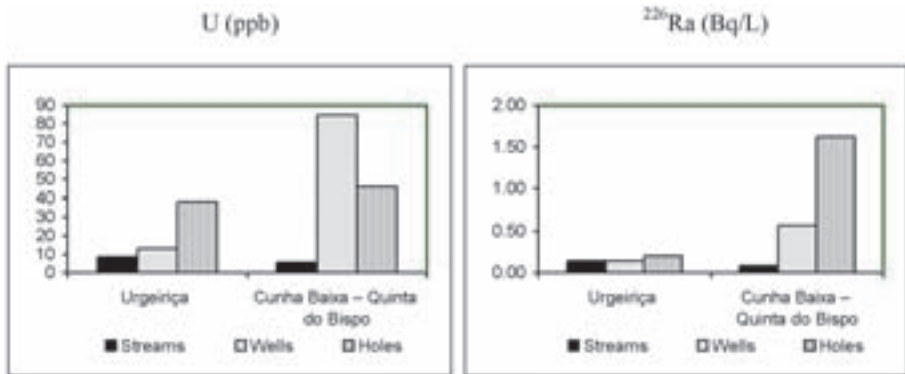


FIG. 3. Uranium and ^{226}Ra averages measured in waters from Urgeiriça and the former Cunha Baixa and Quinta do Bispo mines (first sampling campaign June 2000).

The variability in the activity of each radionuclide in the same environmental compartment is quite high, as already observed for the hydrological compartment .

TABLE 4. MEAN AND STANDARD DEVIATION OF RADIONUCLIDE ACTIVITY MEASURED IN STREAM SEDIMENTS AND SOILS COLLECTED AT THE FORMER URGEIRIÇA AND CUNHA BAIXA-QUINTA DO BISPO MINES

	Area	^{210}Pb (Bq/kg)	^{226}Ra (Bq/kg)	^{230}Th (Bq/kg)	^{238}U (Bq/kg)
Stream sediments	Urgeiriça	811 ± 904	282 ± 200	3465 ± 4912	1088 ± 1006
	Cunha Baixa- Quinta do Bispo	212 ± 158	146 ± 104	82 ± 116	183 ± 212
Soils (layer A)	Urgeiriça	303 ± 151	257 ± 133	978 ± 1182	1297 ± 1604
	Cunha Baixa- Quinta do Bispo	695 ± 692	687 ± 692	1081 ± 1541	764 ± 833
Soils (layer B)	Urgeiriça	342 ± 179	271 ± 143	1244 ± 1425	1860 ± 1620
	Cunha Baixa- Quinta do Bispo	382 ± 508	400 ± 554	381 ± 606	432 ± 608

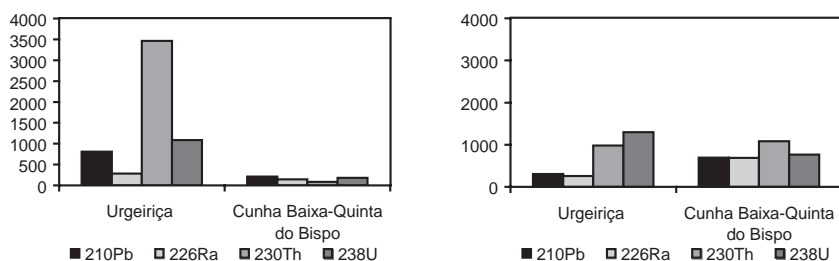


FIG. 4. Radionuclide activity concentrations (Bq/kg) in samples of stream sediments (left) and samples of the more superficial layer of soils (right) collected in the studied areas.

5. DISCUSSION

The variability of the radionuclide activities observed for each environmental compartment results from a mixture, in each dataset, of two sub-populations. One of these consists of samples with a chemical signature changed by mine workings, while the other includes samples that only contain the variability imposed by geology [2, 3]. The separation of the two sub-populations

applying a mathematical technique, i.e. discriminant analysis, allowed a precise identification of the areas contaminated by the mine workings [1].

The average U content and ^{226}Ra activity in waters that percolate the underground mines of Urgeiriça and Cunha Baixa, as well as the tailings located close to the old mines, indicate a high potential for remobilization of the radioactive elements. However, for the same radionuclides significant variations can be detected between water samples collected at different mining sites.

For example, the groundwaters in the Cunha Baixa area contain higher amounts of ^{226}Ra compared to those that circulate around the Urgeiriça mine, which is an indication that the leach rate from the geological materials is higher in the former area. However, the ^{226}Ra activity is higher in the geological materials of the Urgeiriça mill tailings, namely in unit B, than in the solid phase of the Cunha Baixa tailings [3]. Despite differences in texture and composition it is possible that the ^{226}Ra is bound in the mineral matrix with a variable degree of resistance to leaching.

Other factors can also be considered to explain the different pattern observed in the two mining areas, such as the effectiveness of the wastewater control measures and the illegal use of wastewaters from the mine or from seepage in tailings. It has already been mentioned that the procedures followed in the wastewater treatment plant of Cunha Baixa are able to remove a smaller proportion of U compared to the similar procedures used in the Urgeiriça plant. Moreover, it has already been mentioned that farmers pump treated water from the Cunha Baixa mine and from seepage in the tailings that is highly enriched in ^{226}Ra to supply their water needs [3]. This practice, carried out only during the dry season, explains the strong seasonal variations in the concentrations of U and ^{226}Ra observed in several sampling points located in the surroundings of that mine [3].

Crop irrigation with contaminated water helps to disperse contaminants in the soil. Therefore, this practice is likely to be responsible for the high activity of the radionuclides in the soils around the Cunha Baixa mine, particularly of ^{226}Ra , which is much higher than the values measured for the same radioisotope in the Urgeiriça area. This can be observed in Fig. 5, where the activity of ^{226}Ra of the contaminated group of samples is plotted as a function of the mining area and determined by discriminant analysis [2, 3]. For comparison purposes Fig. 5 also shows the maximum permissible limit for ^{226}Ra proposed by the US Environmental Protection Agency (EPA) [5]. For the topsoil (layer A), EPA proposes a value of 185 Bq/kg above background activity, which is assumed in this study to be 115 Bq/kg [6]. Only two samples from Urgeiriça have ^{226}Ra activities slightly higher than the maximum permissible

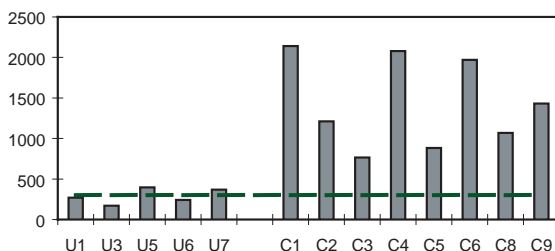


FIG. 5. Comparison of the ^{226}Ra activity concentration (Bq/kg) measured in the contaminated soil samples (layer A) of the Urgeiriça (U), and Cunha Baixa–Quinta do Bispo (C) areas with the maximum allowance limit proposed by the EPA (dashed line). The assumed background activity was 115 Bq/kg [6].

limit, while in Cunha Baixa all samples exceed that limit, being up to 7 times the reference value proposed by the EPA.

An inverse situation is observed for the radionuclide concentrations in stream sediments, which are much higher in Urgeiriça compared to the Cunha Baixa–Quinta do Bispo area. It is suggested that the sources of this difference are the Urgeiriça mill tailings. These tailings consist of fine to very fine grained materials that are subject to mechanical erosion that is able to remove and transport the particles to the nearby watercourse (Ribeira da Pantanha). Several rather small dams exist in this watercourse that can cause retention of contaminated sediments and chemical compounds transported in suspension. Thus the dams can act as settling basins, helping to remove part of the sediment load from the water [2].

6. CONCLUSIONS

In this study, the distribution of various radionuclides in and around three former uranium mine sites in central Portugal was compared. The samples collected from waters, stream sediments and soils reveal some major differences, namely related to the behaviour of ^{226}Ra . The groundwaters that percolate the underground mine of Cunha Baixa and the surrounding tailings release this radionuclide more easily than the other areas studied. In addition to textural and compositional variations it is suggested that the mineral matrix in which the radionuclides are bound can also be responsible for the observed variations.

The varying effectiveness of the procedures used in the wastewater treatment plants and the illegal use of contaminated water from the treatment

plants, or seepage from the tailings, can explain the high ^{226}Ra activity found in soil samples from the Cunha Baixa area compared to the other mining areas. The implications of this finding for the food chain need to be evaluated in further investigations.

The mill tailings of Urgeiriça can be the source of the radionuclides, particularly ^{238}U and ^{230}Th , that occur in high concentrations in the stream sediments of the Ribeira da Pantanha. However, the small dams built along this watercourse help to settle the solid contaminants.

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Paper 17

CHARACTERIZATION AND RISK ASSESSMENT OF SITES CONTAMINATED BY NATURAL SERIES RADIONUCLIDES

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Abstract

Radioactive isotopes are widespread in the environment. In addition to the nuclear industry, oil and gas extraction, the mining of ores, ore processing, fertilizer and abrasives production, military activities and power generation from coal, all contribute to the annual radiological dose received by members of the public. However, whereas routine discharges from nuclear licensed sites are carefully monitored, the fate of natural series radionuclides concentrated by industrial processes is less well known. The nature of these activities dictates that radioactive contamination is often dispersed over large areas and accompanied by elevated concentrations of organic contaminants, heavy metals or both. This presents a severe challenge to regulators and those tasked with implementing a remediation strategy for such complex sites. Assessing the risk associated with ingestion or inhalation of radioactive contaminants relies on detailed characterization of the transport medium and potential source sinks for pollutants along the transport pathway. Currently, reliance is placed on empirical sorption or transfer coefficients to account for retardation and uptake, respectively. Models of this type are difficult to justify. They fail to describe the processes occurring and it can be shown that the outcome of such risk calculations is not necessarily conservative. It is essential to differentiate pollutants in terms of their mineralogy, chemistry, morphology, solubility and isotopic composition, as only this provides an adequate basis for future decision making. Procedures used to devise remediation strategies are illustrated by reference to recent case studies.

1. INTRODUCTION

Despite being the main focus of concern, the nuclear power generating industry is a relatively minor contributor to the annual radiological dose received by members of the public. Routine discharges from nuclear licensed sites are carefully monitored and their potential effect on terrestrial and marine ecosystems is subject to intense scrutiny. In contrast, the fate of natural series radionuclides concentrated by industrial processes is less well known, even though they often occur at levels that cannot be disregarded on health or environmental grounds. The problems caused by naturally occurring radioactive material (NORM) during oil and gas extraction have been known for many years, e.g. Ref. [1]. Mining of metalliferous [2] and non-metalliferous [3] ores, together with ore processing, also generates large quantities of radioactively contaminated material. The fertilizer/phosphate industry, abrasives production and power generation from coal are affected to varying degrees. The nature of these activities dictates that radioactive contamination is often accompanied by elevated concentrations of organic contaminants, heavy metals or both. This presents a severe challenge to regulators and those tasked with implementing a remediation strategy for such complex sites.

While awareness of the problems caused by NORM is increasing, development of a suitable framework for radiological protection still lags some way behind. There are several reasons for this, including underestimation of the perceived risks, cost and the difficulty in harmonizing procedures across national borders and different industrial sectors. Attempts are now being made to ensure greater consistency, for example within the recommendations of Ref. [4]. Nevertheless, and despite the very high activity levels that may be encountered, regulations governing exposures to NORM are generally much less stringent than those for comparable activities of artificial radionuclides.

Reviews carried out on behalf of the European Union [5, 6] serve to highlight the extent of the problem but provide few, if any, details regarding the characteristics of the material itself. This is unfortunate since assessing the risk associated with ingestion of radioactive contaminants relies on detailed knowledge of the transport medium, usually groundwater, and the solubility, bioavailability and toxicity of the contaminant, all of which are species dependent. Similarly, the inhalation of radioactive contaminants in fine, particulate form is known to pose occupational hazards across a wide range of industries and, in rare cases, a risk to public health. However, the severity of the hazard depends on retention by specific receptors and the clearance rate from the body, again dependent on speciation. In the past, representation of such processes has reduced to empirical 'sorption' or 'transfer' coefficients. Models of this type are becoming increasingly difficult to justify as, not only do they fail

to adequately describe the processes occurring, it can be shown that the outcome is not necessarily conservative. The characteristics of the more common materials in NORM from selected industries are described below in terms of mineralogy, chemistry, morphology, solubility and isotopic composition. Assumptions employed in calculating health detriment are explained by reference to actual and potential sources of radioactive contamination in the United Kingdom and Germany. These case studies are used to illustrate the limitations of conventional models and the factors that need to be considered in arriving at a more comprehensive assessment of radiological risk.

2. WASTE ARISING AND ACTIVITY LEVELS

The amount of NORM generated in the EU states was reviewed by Martin et al. [5]. These authors estimated arisings to be at least several hundred thousand tonnes per annum, dominated by ash from coal fired power stations and the residues of phosphate processing, metal smelting and hydrocarbon production. A more detailed study of the German situation conducted by Leopold et al. [6] suggested greater volumes and even assigned wastes to individual producers (though in many cases the operator of the facility in question was reluctant to provide quantitative information). On the basis that each tonne of raw phosphate generates 1.5 tonnes of phosphogypsum, potential annual arisings in Germany alone could amount to more than 400 000 tonnes. Schmidt et al. [7] addressed radioactivity in waste material specifically from phosphate ore processing, comparing findings and regulations in Europe with those in the United States of America. It emerges that such residues constitute a significant problem worldwide.

A feature common to most of the reviews carried out to date is the generally low specific activity accorded to industrial NORM, which often appears to be at odds with the experience of workers in the various industries affected. This discrepancy is illustrated in Table 1 where values quoted as 'typical' in Ref. [5] are compared to those found during recent investigations in the UK and Germany. Clearly, assuming unrealistically low activities would lead to a serious underestimation of the radiological hazard.

Specific examples of NORM contamination in non-metalliferous mining, natural gas extraction and phosphate production are discussed in the following sections. Different approaches to the problem have been devised on a case by case basis, reflecting the extent of the contamination and local factors. These are also described as they may provide guidance to workers in other fields.

TABLE 1. 'TYPICAL' (a) AND MAXIMUM (b) ACTIVITIES FOUND IN INDUSTRIAL NORM RESIDUES. THE HIGH ^{210}Pb CONTENT (c) RELATES TO DUST FROM THERMAL PROCESSING

Industry/Source	Activity (Bq/g)		
	^{238}U	^{226}Ra	^{210}Pb
Mining			
Martin et al. (1997) [5] (a)	1–10	1–20	1–20
Leopold et al. (2002) [6] (b)	<30	<20	<20
Read et al. (2004) [3] (b)	270	6100	7700
Gas extraction			
Martin et al. (1997) [5] (a)	-	200	50
Leopold et al. (2002) [6] (b)	-	1000	320
Ceccarello et al. (2004) [8] (b)	-	36	7550
Phosphate production			
Schmidt et al. (1995) [7] (a)	0.5	3	1.3
Leopold et al. (2002) [6] (b)	1	0.8–1.25	1000 (c)
Beddow et al. (2004) [9] (b)	9	11	12

3. CASE STUDY 1: NON-METALLIFEROUS MINING

The mineralogical constituents of NORM vary, depending on the process being considered. Radionuclides present in water, rocks or sediments at very low mass concentrations may display high selectivity for certain mineral lattices. The best known example is the co-precipitation of radium isotopes in barite (BaSO_4). This almost insoluble phase is the dominant scale mineral affecting oil and gas installations [10], and is a major contributor to radiological dose in uranium mill tailings [11]. It has recently been observed in pipe work, centrifuges and other plant used in the refining of china clay (kaolinite). The characterization and treatment of these deposits as part of an integrated waste management strategy is described in Ref. [3].

The china clay deposits of Cornwall and Devon are the largest in the world and have been mined for almost 250 years. High grade uranium ores are known to occur close to the major kaolinite bodies, and consequently there is a risk that radioactive isotopes may become entrained with the clay during mining and concentrated by subsequent chemical treatment. Once removed

from the pit face, the raw material is subjected to a number of physical separations to increase the kaolinite component and to achieve the desired grain size. Sodium hydroxide as a dispersant and sulphuric acid are added at several stages during this process, the effect being to release barium and trace constituents from the mica, feldspar and accessory minerals. Bleached clays receive additional dosing with acid and a reducing agent (sodium dithionite) to remove iron. The large volumes of raw material processed means that there may be a substantial inventory of even ultra-trace components passing through the plant.

Although processing has been carried out for many years, radioactive scaling was first discovered in 1996. Subsequent investigations confirmed that the problem was widespread and a comprehensive sampling and characterization programme was initiated [3]. The scale, consisting almost entirely of barite and kaolinite, occurs as thin, discontinuous coatings reaching up to 100 mm in thickness (Fig. 1).

The proportion of barite in individual samples ranges from <1% to >90% and correlates directly with the activities of the principal radionuclides, ^{226}Ra and ^{228}Ra (Fig. 2). For any given stage of the refinement process the degree of selectivity shown by barite for these isotopes is constant. Thus, normalizing activities for Ba content allows maximum radium activities to be calculated. Regulations governing the disposal of such wastes require that the material be 'substantially insoluble', a condition that is satisfied in the case of radium. However, the proportion and solubility of ^{210}Pb varies with the age and

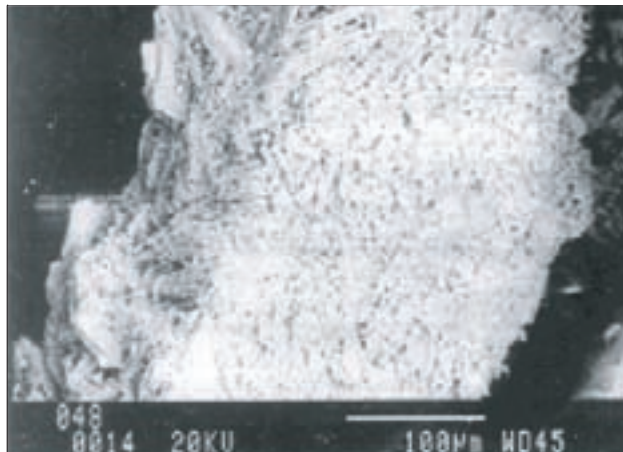


FIG. 1. Scanning electron image of barite scale from clay processing.

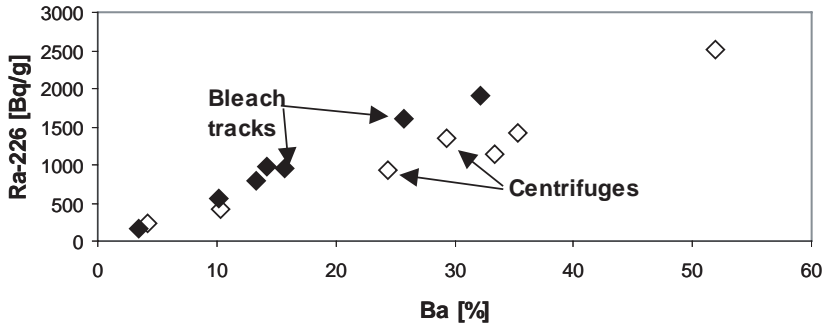


FIG. 2. Correlation of ^{226}Ra activity with Ba content in scales.

treatment history of the deposits [3]. This has a bearing on both disposal practices and potential health detriment. Therefore, sequential leach tests were performed to assess the stability of the scale when exposed to progressively more aggressive reagents.

Decontamination trials were undertaken and a decision made to adopt ultra high pressure water jetting as the preferred removal method for scale. A state of the art facility was constructed in Cornwall and entered service in 2003. Decontamination gives rise to slurry comprising scale, associated minerals, corroded metal and large quantities of water. Extensive research to define a suitable immobilization matrix identified a highly durable cemented product with excellent leach resistance. Full scale specimens have now been prepared and, following regulatory approval, have been consigned to an appropriate waste facility.

4. CASE STUDY 2: NATURAL GAS EXTRACTION

During oil and gas extraction, mixing of formation waters coupled with changes in temperature and pressure often causes scaling in tubing and surface installations. The most common scale mineral encountered in oil production is barite, which incorporates radium isotopes by ionic substitution, as noted above. Other phases occur in gas extraction and these tend to be characterized by high levels of ^{210}Pb (and ^{210}Po), owing to the sublimation and deposition of volatile compounds. Two such deposits have been investigated recently [8, 12].

The Altmark region in Sachsen-Anhalt, Germany hosts major gas production fields, where wells into the Lower Permian reservoir rocks can reach a depth of 3500 m. The gas itself contains mercury at concentrations of up to 4 mg/m^3 . Kaemmel et al. [13] described crusts of lead and mercury rich scales

on pipe work but, at this time, it was not known that the material was also highly enriched in natural series radionuclides. A re-examination of the deposits, termed ‘altmarkite’ (HgPb_2), has shed light on this issue, as described in more detail in Ref. [8].

Macroscopically the deposits are massive and apparently amorphous with a pronounced metallic lustre (Fig. 3). Under the microscope, the material displays a degree of crystallinity but is very heterogeneous, particularly on oxidized surfaces; amalgams with compositions ranging from 5.1–94.3 to 90.6–8.9 (Hg–Pb wt%) have been found. The Pb–Hg deposit shows significant activity with regard to all of the principal chain radionuclides with the exception of radium. ^{210}Pb is the main radionuclide present, with an activity exceeding 4000 Bq/g. Impurities, mainly $(\text{Ba},\text{Sr})\text{SO}_4$, are associated with microscopic regions of radium accumulation [8].

In bulk analyses the mercury content ranges between 20 and 40%, sufficient to constitute an economic resource in its own right. Removal of mercury from the HgPb_2 is technically feasible, although the materials remaining are still hazardous. A bespoke plant has been constructed in Leipzig to recycle the mercury, after which residual scales are encapsulated in a geopolymer containing an alkali activated aluminosilicate matrix [14]. The matrix has projected long term stability and has been accepted by the German regulators for the disposal of NORM wastes in municipal landfills.

Similar deposits, rich in ^{210}Pb , have been found on gas production equipment from the onshore Wytch Farm field, UK [12] and the Netherlands [7], implying that the incidence of radioactive lead deposits is widespread in Western Europe. Detailed mineralogical, chemical and radiochemical analysis of the former is ongoing. However, preliminary data indicate a predominance

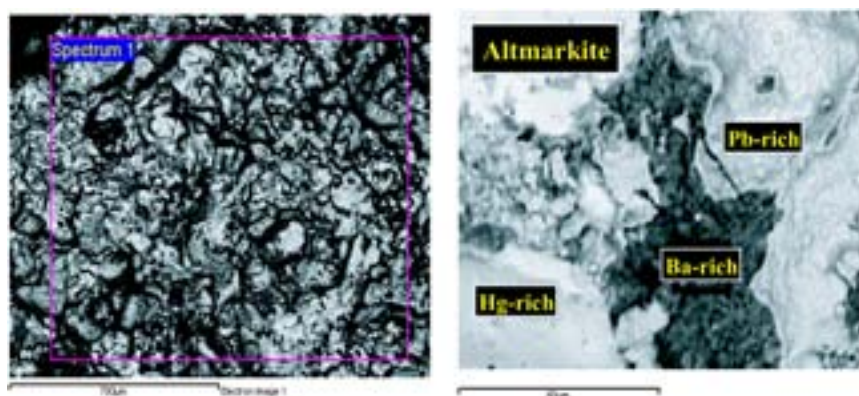


FIG. 3. SEM image and elemental maps for ‘altmarkite’ (HgPb_2).

of metallic lead and, therefore, the scales are effectively insoluble. This would seem to justify current disposal practices that involve direct re-injection of the scales into the sub-surface.

5. CASE STUDY 3: PROCESSING OF PHOSPHATIC ORES

A database of sites affected by NORM is being compiled in order to quantify the extent of radioactively contaminated land in the UK. As part of this study, a phosphoric acid processing works was selected for more detailed mineralogical and chemical characterization of the waste generated [9].

The plant, located on the northwest coast of England, produced phosphoric acid from 1954 until 2002. The company originally imported sedimentary phosphate ore (carbonate-fluoroapatite pellets) containing approximately 1490 Bq/kg uranium, from Morocco and Israel. This ore was reacted with sulphuric acid to produce an impure 'green acid', which was then further purified to produce a food, technical and salt grade phosphoric acid. The waste products, comprising phosphogypsum slurry and an acid waste stream, were discharged into the Irish Sea under UK authorization [15]. On-site manufacture of phosphoric acid from ore was abandoned in 1992 and instead 'green' phosphoric acid was imported from Morocco and Mexico and purified at the works. A plant was constructed to treat the liquid waste with quick lime (CaO) to produce a solid (termed 'Ufex') consisting of calcium sulphate and calcium hydroxyapatite, which could then be landfilled on-site. Discharge to the sea was discontinued.

During demolition of the old workings it became apparent that many of the installations were contaminated with radioactive scales (Fig. 4). These materials required characterization to establish whether they were suitable for on-site disposal.

The scales constitute a much more complex mineralogical suite than the deposits described previously. They predominantly comprise fluorides (e.g. Na_2SiF_6 , NaMgAlF_6 , $\text{CaMg}_6\text{Al}_2\text{Si}_6\text{O}_{20}\text{F}_4$), calcium sulphate (gypsum, anhydrite and hemihydrate) and a mixed assemblage of fluorides and phosphates (e.g. $[\text{Mg,Ca,Fe}]_2(\text{PO}_4)\text{F}$, $\text{Ca}_2[\text{Mg,Fe}](\text{PO}_4)_2 \times \text{H}_2\text{O}$), related to the various processes occurring during impure acid production and purification [9]. As expected, ^{238}U and its daughter ^{234}Pa were found in installations where the process stream was rich in fluorides and/or phosphates, whereas high levels of ^{226}Ra were found in association with precipitates of calcium sulphate. It was noted that several materials from the acid purification plant had ^{210}Pb activity levels in excess of parent ^{226}Ra .



FIG. 4. Scale on pipe work from phosphoric acid processing.

Maximum activity levels were found for the main isotopes ^{238}U , ^{226}Ra and ^{210}Pb are 9, 11 and Bq/g, respectively. These values approach the current thresholds for landfill disposal of insoluble solid wastes in the UK, though it is noted that they reflect the highest rather than average levels found.

6. CONCLUDING DISCUSSION

The examples presented above highlight the importance of establishing mineralogical associations for the radioisotopes present in NORM. This information is a prerequisite for determining their solubility in different media and hence appropriate treatment and disposal procedures.

In developing preferred management options for industrial NORM in clay manufacture, several assessments were carried out to underpin the decision making process [3]. These included a best practicable environmental option study to support the technology selection process; an environmental impact assessment to determine the potential effect of the proposed refurbishment plant on the local environment; a radiological risk assessment and a 'HAZOP' risk assessment. Reliable data could not have been obtained from these assessments without prior knowledge of the mineralogy, chemistry and uranium series disequilibrium characteristics of the material.

Although barite itself is insoluble, microcrystalline particles are still mobile and the material presents a hazard if inhaled. This, together with the greater solubility displayed by ^{210}Pb in recent deposits, necessitated immobilization in a durable matrix. Similar considerations apply to the lead–mercury

scales found in German gas production facilities [8]. Conversely, fixation of the same nuclide in metallic form permits direct disposal, subject to activity levels meeting regulatory approval. Finally, the complex assemblages found in scales generated by phosphoric acid processing suggest that further work is needed to assess their stability in groundwater solutions and likely clearance rates if inhaled or ingested.

ACKNOWLEDGEMENTS

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Paper 18

LESSONS LEARNED FROM ACTIVITIES CARRIED OUT UNDER THE BUHOVO TAILINGS POND RECONSTRUCTION PROJECT IN BULGARIA

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Abstract

In 1947, the ‘Metallurgy’ uranium processing plant began operation in Buhovo, near Sofia. Until 1990 the plant processed 10 million t of ore from various uranium deposits throughout the country. Tailings arising from the uranium recovery process were placed in two ponds, covering a 24 ha area close to Monastirsko river. Disposal took place in a basin enclosed by an earth dam. A great quantity of liquids remained above the sandy tailings, creating a severe risk in case of dam failures under seismic conditions because of the potential for liquid runoff and ensuing downstream contamination of the Monastirsko river. To strengthen the dam, a 30 m wide additional berm, extending to an elevation of 678 m, was planned and built. It required 700 000 m³ of clay soils and 60 000 m³ of gravel to be used as a filter for drainage purposes. Construction work was carried out by DEC (a Belgian contractor) between March 2000 and August 2001. INITEC, acting as the architect/engineer, supervised the work under FIDIC (Fédération Internationale des Ingénieurs-Conseils) conditions and Bulgarian law. The purpose of the paper is to describe the lessons learned during this project, as well as the INITEC role as architect/engineer with active involvement in design modifications aimed at improving the performance of the project.

1. PROJECT BACKGROUND

Buhovo is a small town some 25 km northeast of Sofia, where a uranium milling plant was in operation between 1947 and 1990, processing some 10 million t of ore from various uranium deposits, using different techniques for hydrometallurgical extraction of the uranium ores. The mill used a variety of chemical reagents, including sulphuric acid, carbonates and bicarbonates, for extracting the uranium, manganese ore, potassium permanganate or nitrates as oxidizers of the uranium, organic substances as flocculants, and lime, kerosene, as well as other reagents necessary for the process of extraction and concentration of uranium, until the final product, ‘yellow cake’, was obtained.

During the early period of operations between 1947 and through 1956, tailings were discharged directly into the Monastirsko river, resulting in a contaminated riverbed area of about 118 ha. Tailings generated from 1957 to 1990 were discharged into two tailings ponds. The older one, with an area of 24 ha, was relatively well compacted and partially recultivated once full, while the new pond still contained large quantities of radioactive liquids at the start of the remediation project (Fig. 1).

To improve the stability of the dam, a two stage project was developed under the umbrella of PHARE funds. In 1997 stage I of this project started with the partial strengthening of a land slide below the surface water diversion channel, cleaning of this channel and construction of an emergency overflow system. This paper deals mainly with stage II, the scope of which is described in the following.

2. STAGE II: RECONSTRUCTION OF THE BUHOVO TAILINGS POND

This project started in 2000 and included the following main activities:

- (a) Strengthening of the tailings pond dam by addition of a downstream berm composed of a drainage/filter layer of gravel (60 000 m³) placed over the

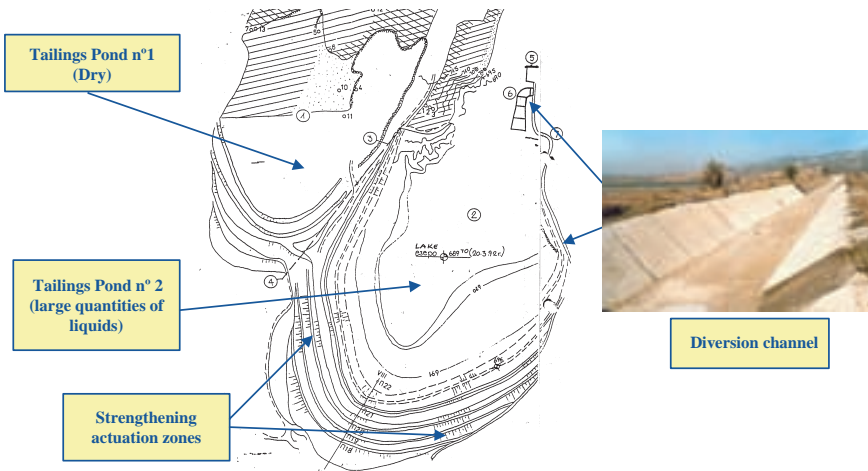


FIG. 1. The Buhovo site.

existing slope and covered by a layer of compacted clay ($700\,000\text{ m}^3$), as shown in Fig. 2;

- (b) Construction of a downstream drainage piping system to collect drainage water from the dam and to conduct this water to a 500 m^3 reinforced concrete underground reservoir;
- (c) Construction, testing and commissioning of a pumping station, complete with automatically controlled pumps and valves, located in a building constructed on the reservoir roof slab;
- (d) Construction of a pressure pipeline to return the drainage water collected in the reservoir to the pond;
- (e) Connection of the pumping station to the local electricity supply network, including construction of a new transformer station, $3 \times 3 \times 7\text{ m}$ in size, and installation of a new 400 kVA , $20/0.4\text{ kV}$ transformer;
- (f) Remedial work to the slope above the tailings pond, where there has been a small landslide; The work consisted of:
 - (1) Construction of a drainage system on the berm at $+697.5\text{ m}$, with outlets;
 - (2) Excavation of existing material;
 - (3) Placing and compacting part of the excavated material and/or clay from the borrow pits to form embankments;
- (g) Construction of a barrage wall upstream of the gully to the tailings pond to divert clean surface water through the existing channel (remaining work from stage I).

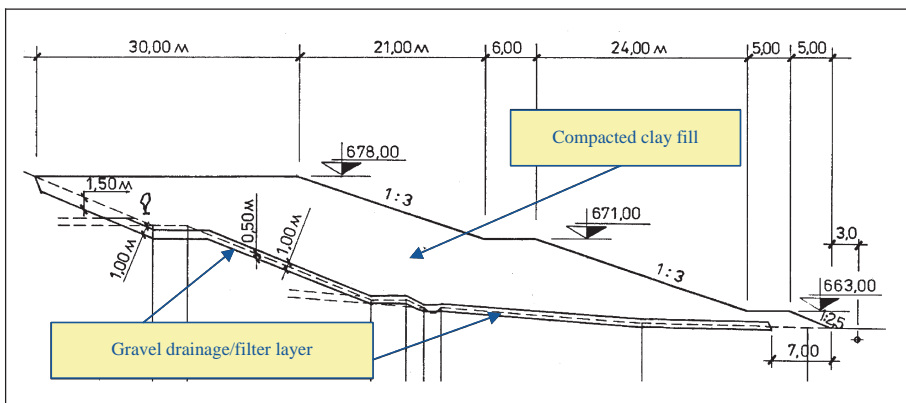


FIG. 2. Tailings dam strengthening plan.



FIG. 3. The 500 m³ underground reservoir.

According to FIDIC conditions and Bulgarian law, INITEC supervised the work carried out by DEC.

Several technical and administrative issues arose during the work, some due to factors that were not anticipated during the design phase, and others having their origin in a new Bulgarian law regulating supervision of civil works that incorporates a complex system for approval of all phases of the work. Finally, the project had to tackle a problem derived from the lack of budget allocated for the purpose of testing and operation of the active parts of the project (pumping station).

3. MAIN TECHNICAL ISSUES

A total of 40 variation orders were analysed and approved by INITEC to cope with the inevitable changes to the original provisions of the project and identified during the actual construction work. The main changes were:

- (a) Since clay soils were used to strengthen the dam, with a weak erosion protection performance, an extended area of the new clay fill was affected

by surface water erosion after the first rains, and large gullies were formed due to the high velocities of the runoff rainwater (Fig. 4). Also, the project does not include any provision for an effective surface water erosion cover. To prevent the formation of new gullies, a combination of different slopes was adopted on the top and slopes of the clay fill, in order to avoid points of high water velocities in the external surface of the clay fill.

- (b) As all drainage through the dam was very acid, with a high content of sulphates, all concrete collection piping was changed to PVC. Also, the pressure pipeline designed to return the water to the tailings pond was changed from steel to HDPE to avoid corrosion. Finally a sulphate resistant concrete was used for the structure of the reservoir, and a plastic liner was put into place to cover its internal surfaces.
- (c) A barrage wall was also added to the project with the aim of diverting all Monastirsko gully surface runoff water, reducing to a minimum the entrance of clean water into the tailings pond, so that the process of natural tailings dewatering would not be compromised.

After much infiltration through the base of the reservoir formed by the dams was observed, a study was conducted to assess the capability of this structure to effectively collect the gully surface runoff.



FIG. 4. Effects of the first rains on the clay fill.

As result of this study, INITEC decided to inject cement into the base of the dams to create a screen that prevented such leaks, ensuring the effective collection of all runoff surface waters and their conduction through the lateral diversion channel.

4. ADMINISTRATIVE ISSUES

A new law that regulates the supervision of civil works with a complex system of approval procedures and cross-responsibilities between local and state authorities went into effect almost at the same time as the start of the project, having a great impact on the final documentation of the project and resulting in a substantial delay of the effective conclusion of the project.

5. FUNDING FOR THE OPERATION OF THE PUMPING STATION

At the time of testing and operation of the pumping station, many problems arose in connection with the funds required for electricity supply for the operating system, due to the site owner's economic situation.

A great risk of possible damage to the equipment had to be assumed because of the delay in starting the operation of pumps and their automatisms. To avoid this sort of problem it would be very expedient to include in the overall project budget the funds required during a reasonable period of time for the operation of any active element incorporated in the remediation design, so that the overall objectives of the project are not jeopardized.

6. CONCLUSIONS

Some technical problems could have been avoided during the design phase, but as is usual in any such work they were satisfactorily solved during the construction period. Only one recommendation could be made: the use of an erosion protection cover while the revegetation of slopes is established, as indicated in Fig. 5.

On the other hand, any effort to simplify the administrative steps for the approval of construction works turns out to be very useful when considering the objective of performing projects in a cost effective manner.

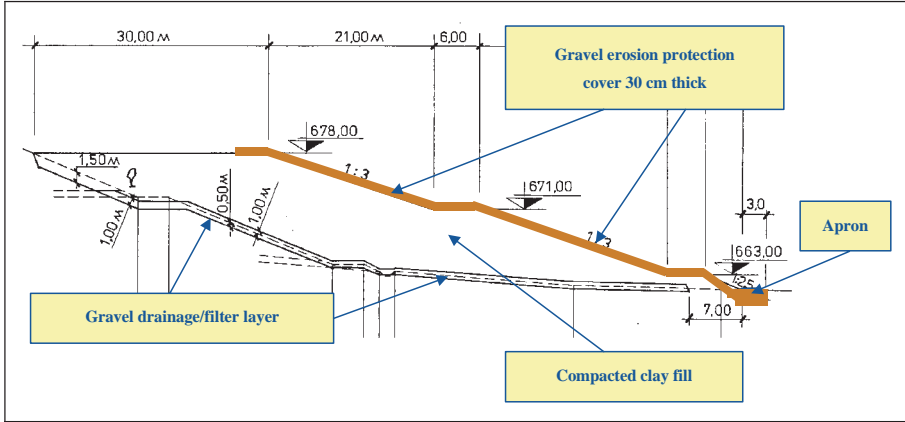


FIG. 5. Erosion protection cover.

The preferred remedial action for this kind of site should always be based on passive designs, avoiding as much as possible the use of active systems such as the pumping station included in this project.

Nevertheless, as was done in this case, temporary solutions may be adopted for the necessary control of contaminated waters until the dewatering of the tailings pond is complete or sufficient to allow the construction of an adequate cover of the impoundment.

In such cases, a careful study of operating costs should be performed, so that the necessary funds required over a well defined period of time may be included as part of the overall project budget, so as to ensure that the project's objectives can be achieved.

Paper 19

URANIUM MILL TAILINGS PILES REMEDIATION: REMEDIAL CLEANUP ACTIONS VERSUS LONG TERM STEWARDSHIP

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Abstract

An important objective of the decision making process for the cleanup and remediation of inactive uranium mill tailings piles is to achieve an adequate balance between the scope of the post-closure cleanup remedial actions and long term stewardship needs, obligations and associated costs. This requires evaluation of both the actual technical and funding feasibility of the proposed remedial actions, and of the feasibility of long term stewardship from the technical, social and cost effectiveness points of view. While it is recognized that long term stewardship should not be considered as an alternative to cleanup, the fact is that it is not always possible to reach a green field end-state for a contaminated site, due to factors such as technical feasibility, worker health and safety, collateral ecological damage and costs. Therefore, in most situations it is not possible to entirely eliminate the need for long term stewardship. The extent to which a remedial action achieves unrestricted use drives the long term stewardship needs and obligations. The paper presents a discussion of the provisions included in the design and construction of the Andújar uranium mill remedial action plan, focusing on the requirements for long term stewardship, aimed to ensure adequate performance of the tailings enclosure cell by means of long term surveillance and maintenance plans. Several key issues and considerations relevant to this site are also discussed in the paper: They cover aspects such as land control and property, public information management, funding and financial provisions, agency or authority responsible for long term stewardship, local, regional and state authorities involved.

1. INTRODUCTION

The tailings generated during the extraction of uranium from ores and the production of U concentrate (yellow cake) are characterized by their very low specific activity, corresponding to the long half-life of the naturally occurring radioisotopes ('NORM'), such as ^{226}Ra and ^{230}Th , and by their very large inventory in terms of mass. They are usually disposed of in large human-made or natural impoundments, closed by perimeter walls or dikes.

The main radiological risks associated with these ponds, when they are not remediated or protected, are well known and shown in Fig. 1.

The usual remedial action for these ponds calls for the reconfiguration of dried tailings to ensure their geotechnical stability and the subsequent placement of an engineered cover designed to isolate the tailings and any other material that is disposed of in the impoundment from the surrounding environment, so that radon emanations and rainwater infiltration are kept to a minimum. In this way effective protection of both the surrounding environment and the public living nearby is achieved, provided that the characteristics of the engineered cover are maintained over time (Fig. 2).

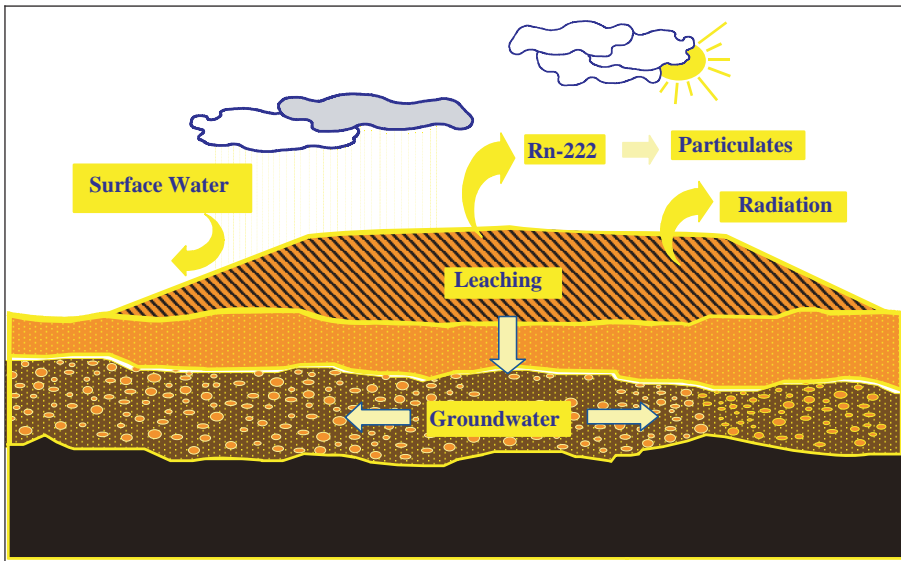


FIG. 1. Main risks from a tailings pile.

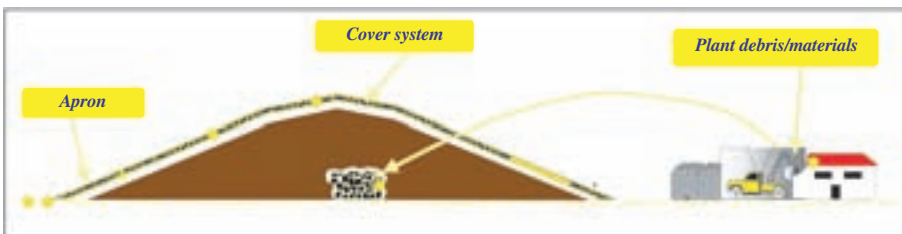


FIG. 2. General approach: Disposal cell.

Cost–benefit analyses are usually performed for different alternatives for the final location of the tailings. In most of the cases, the option of in situ stabilization and disposal is selected. The relocation of the whole tailings inventory to a different disposal site is considered only on very rare occasions, but even in such cases the basic scheme depicted in Fig. 2 is retained.

It is important to note that the current technological approach to tailings management does not permit a residential scenario allowing unrestricted use of the remediated ponds from the completion of the remediation work on.

It is therefore mandatory to establish and implement long term access restrictions to the restored site, regardless of the safety and reliability of the design of the engineered cover. These access restrictions will be effected by, e.g. a governmental control programme that will also include in its objectives maintenance in the future of a memory of the site and of the materials disposed of there.

2. CLEANUP DECISION

As mentioned previously, during the decision making process to be applied for the selection of remediation alternatives for uranium mill tailings ponds, due consideration must be given to the relative efforts and costs associated with both, the site remediation activities and the long term control, surveillance and maintenance of the remediated site, also known as ‘long term stewardship’. In this way, the estimated effort and costs associated with each solution investigated will be the sum of both types of activity.

It must be recognized that both extreme situations, i.e. remediation with no future restrictions, or stewardship requirements and no remediation but trust in an indefinite site custody and control by the relevant authorities, are not feasible. In the first case the reason is a technological one, as explained above. In the second case the alternative is judged unacceptable on the grounds of societal considerations. Therefore, and depending on specific site conditions, the optimum solution will be an intermediate point on the diagonal in Fig. 3.

A typical engineered cover for a tailings pond is shown in Fig. 4. As may be seen here, a first layer of clay is used as a barrier to reduce radon exhalation from the tailings, and as waterproofing to prevent the infiltration of rainwater into the tailings. A second layer of granular material is placed on top of this clay layer to provide drainage for rainwater, directing the water sideways to lateral drainage channels at ground level. The third layer provides protection against wind and water erosion of the lower layers, and may consist of gravel or a layer of vegetated topsoil.

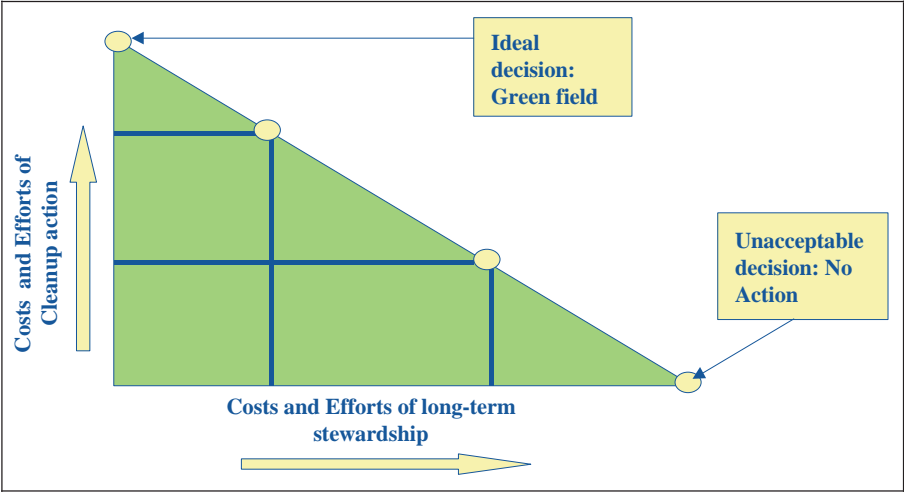


FIG. 3. Cleanup decision vs. long term stewardship.

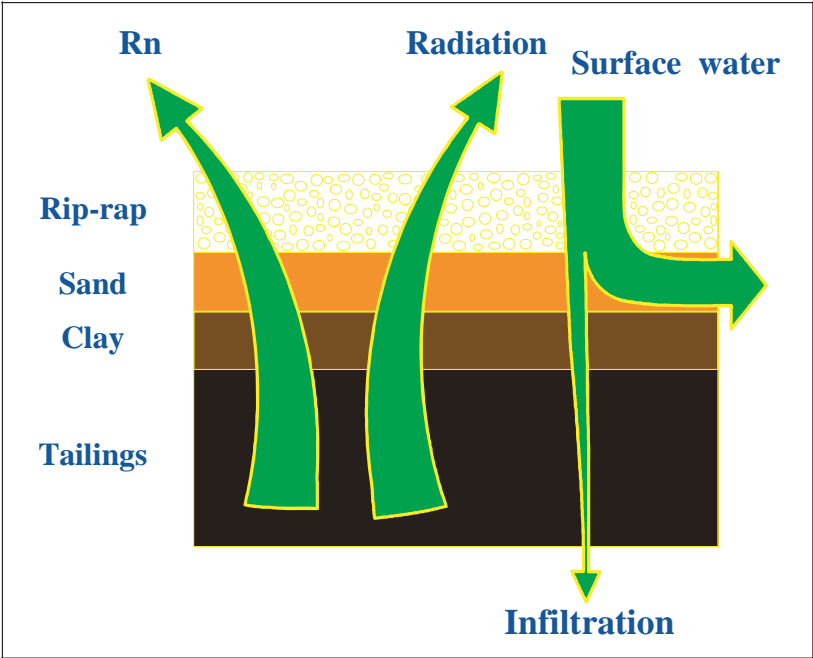


FIG. 4. Multifunctional cover design.

Engineered covers of greater complexity may be required at sites with shallow aquifers. This is the case at the Andújar uranium mill in Spain, where the solution adopted included the extra layers presented in Fig. 5. This approach also reduced to a minimum the long term maintenance requirements for the cover. In general it may be noted that this type of complex cover significantly reduces long term custody requirements, while the simpler type, i.e. that relying on a single vegetation cover for erosion protection, usually leads to much greater long term maintenance and custody requirements, as shown in Fig. 6.

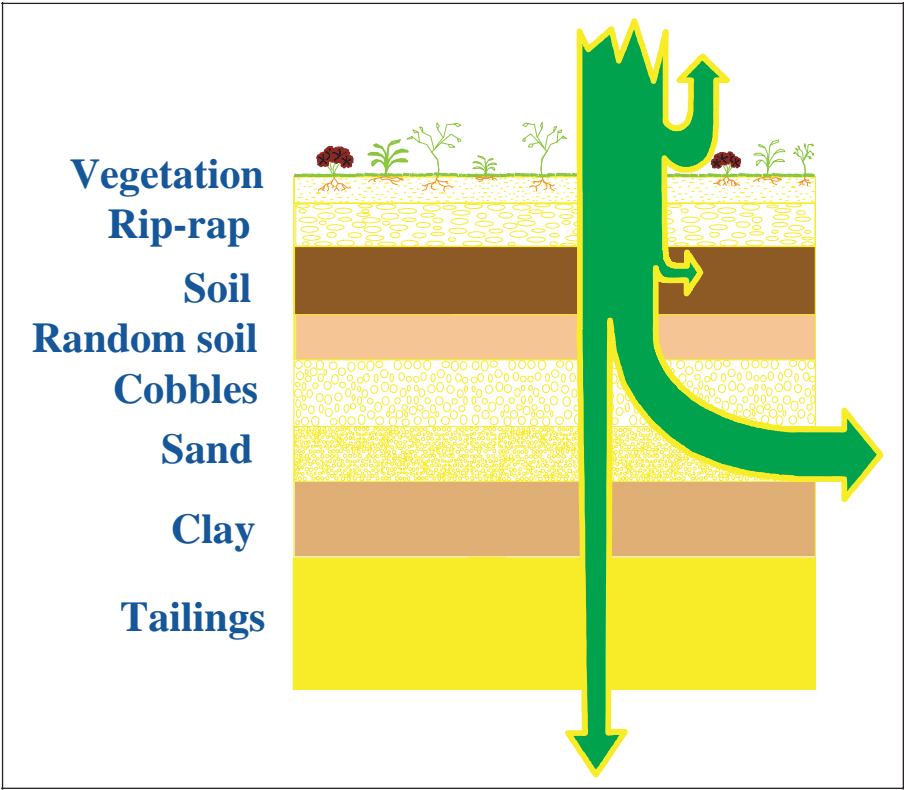


FIG. 5. Complex cover design.

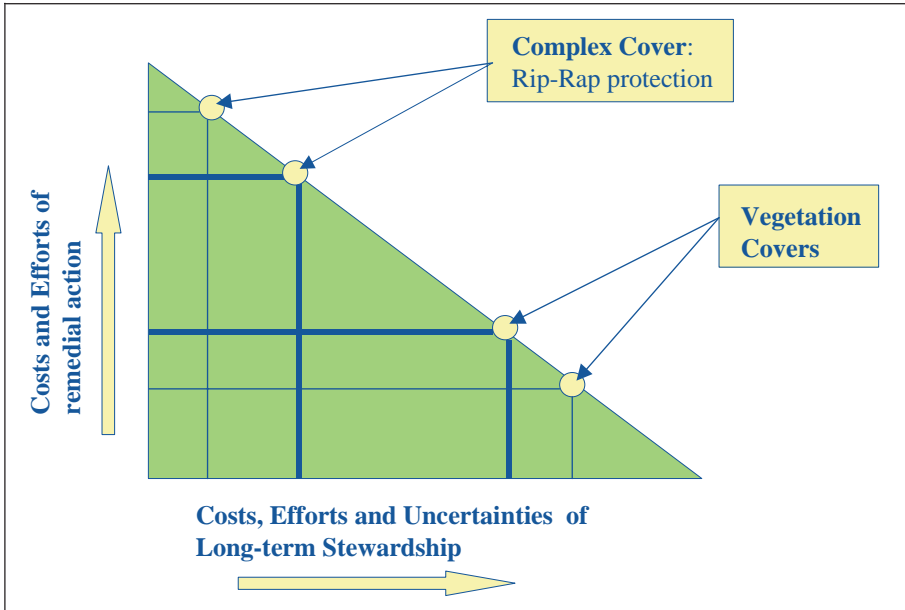


FIG. 6. Cover design versus long term stewardship needs and uncertainties.

3. LONG TERM STEWARDSHIP NEEDS AND REQUIREMENTS

3.1. Demonstration of compliance with design limits

Any remedial action based on isolation of tailings using the disposal cell concept, that is confined by the ground below the pile, an engineered cover and surrounding dikes (cf. Fig. 2), must be subject to a maintenance and surveillance programme over a 5–10 year period after completion of the remedial actions. The objectives of this programme are to:

- (a) Verify compliance with both the ^{222}Rn emission limits from the pond and with the groundwater quality criteria established for the initial transient period;
- (b) Verify the adequacy of the design adopted to maintain the long term integrity and stability of the cell, as well as to evaluate the need to perform maintenance operations, mainly related to the engineered cover.

3.2. Verification of integrity and stability

There will be a critical transient period, for instance due to changes in the hydrological water balance, until a remediated site reaches compliance with the established limits. This period may last for 20 to 30 years. During this period only the overall integrity and stability aspects of the remediated pond should be controlled and verified using simple standard civil engineering surveillance techniques. Examples of these techniques are visual observation for signs of erosion, surveying to detect settling, verification of the presence of markers and access control features, e.g. fences, etc.

The responsibility for these controls, as well as for overall site surveillance and record keeping, can and should be transferred to local civil authorities with the aim of ensuring effective control over the uses and access of the site area and maintaining, for future generations, the memory of the site history and imposed use limitations. They will also be able to perform the required inspections and repairs needed after catastrophic events such as floods, earthquakes, etc.

3.3. Severe environmental risk surveillance period

After a 30–40 year period following remediation, only low cost and simple controls should be in place. They will concern, basically, the keeping of records on design data, results of monitoring and surveillance activities, the characteristics of the pond materials, provide advice and instructions for checking the status, and on repairs of the stabilized pond after the occurrence of severe environmental phenomena.

4. CUSTODY AND SURVEILLANCE PERIODS: PROPOSAL FOR LONG TERM STEWARDSHIP

As stated previously, the use of a tailings isolation and confinement concept that involves high stability runoff water erosion protection elements, e.g. a ‘rip-rap’ layer, allows for a significant reduction of the long term custody and surveillance requirements, which may be reduced to simple low cost, overall integrity checks. Responsibility for such checks may be assumed by the local authorities or they may be easily integrated into existing normal community infrastructures, surveillance and maintenance programmes at the regional or national level; all this at a low cost and with no significant detriment to public safety.

In this context it is important to recognize that the unavoidable uncertainties in the long term evolution of human society make it unwise to assume that a regular flow of funds will be available for these purposes. Therefore the simpler and cheaper the long term control requirements are, the more probable it will be that these controls are actually performed. This is even more relevant when considering time horizons measured not only in decades, but also in centuries.

It is also unwise to contemplate this problem from the perspective of today's opulent western society, with large monetary budgets. In the long term the situation may change for the worse, and powerful private or public entities that would normally be responsible in these matters may eventually disappear as a result of social turmoil or disruption.

Figure 7 presents a proposed long term custody and control strategy where the scope and depth of the verifications will evolve with time in a step-wise manner. The three periods that define this strategy are:

- (1) Compliance period (0–10 years), during which compliance with ^{222}Rn exhalation and groundwater quality limits must be checked regularly.

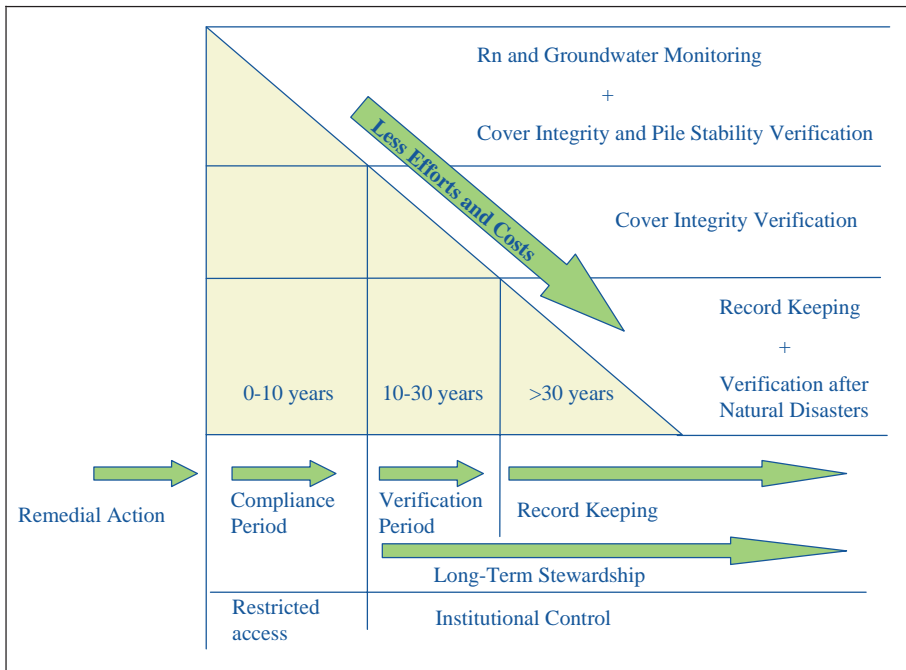


FIG. 7. Proposed scope/periods of stewardship.

Strict controls over access, integrity and geotechnical stability must be in place.

- (2) Verification period (10–30 years), during which surveillance of the cover integrity and the overall stability should be used as indicators of compliance with ^{222}Rn and groundwater limits.
- (3) Institutional control period (>30 years), during which design and past surveillance records, as well as the memory of site origin and use limitations, should be kept. The overall integrity of the remediated site should be verified during and after severe adverse environmental phenomena (e.g. great floods or earthquakes).

With regard to responsibilities, it is clear that those related to the remediation effort and with the compliance period should be borne by the previous site operator or the designated government agency. After the compliance period the ideal situation would be that the responsibilities during the verification and institutional control periods be transferred to the relevant local authorities, which would use funds transferred by the central government for this purpose.

5. CONCLUSIONS

It should be recognized that cleanup decisions for sites affected by uranium tailings piles will always be located in an intermediate point between ‘green field’ and ‘no action’, taking into account cost and efforts to perform a specific remedial action and associated long term stewardship tasks and requirements.

As a green field approach is currently unreasonable, considering the great quantities of materials, usually involving millions of tonnes, and the very low specific activity of long lived radionuclides, long term stewardship measures are unavoidable. A three period approach to long term stewardship after remedial action is completed is suggested:

- (1) Compliance period (0–10 years), under the responsibility of the agency that performs the remedial action to verify that design and regulatory objectives are met, followed by:
- (2) Verification period (10–30 years), under state control, to verify pile stability and integrity, followed by:
- (3) Institutional control period (>30 years), under the control of local authorities, to verify overall integrity and record keeping.

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Paper 20

THE PROGRAMME FOR REMEDIATION OF CONTAMINATED MINE SITES: ITS REGULATION AND FOLLOW-UP IN PORTUGAL

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Abstract

The policy of the Portuguese Government of assuming responsibility for remediating contaminated abandoned mine sites originated in an initiative taken between 1995 and 2001 by the General Directorate for the Environment and the Geological and Mining Institute. It has the aim of assessing and solving the prevailing environmental problems in some of the most contaminated abandoned mine sites in Portugal. On 6 July 2001, through Decree Law No. 198-A/2001, the Government defined the institutional and financial provisions to be adopted for implementing the environmental remediation programme. EXMIN-Industry and Mining Environmental Services S.A., a state owned company, was awarded an exclusive renewable contract on September 5 of the same year for a period of 10 years to implement this programme. Financing of the contract was guaranteed through EU funds under the FEDER programme, up to a maximum of €52 million, to be spent before the end of the year 2006. In the beginning of 2002 a steering committee was nominated and took up its responsibilities, having been delegated a wide range of powers. At the same time the Ministers for Economic and Environmental Affairs delegated wide ranging powers to a technical evaluation subcommittee. The strategic definition of the targets of the old mine site remediation programme, which remains under the direct responsibility of the relevant ministers, highlights the importance of public health and safety and the social and economic development of the regions concerned. The information already gathered indicates that in total around 170 old mine sites require remediation. The time limit of 2006 for the availability of EU funds needs to be taken into consideration for the development of the remediation strategy. The specific situation of the old radioactive ore mines is described, as well as the status of the programme.

1. HISTORICAL BACKGROUND

Raising international and State administrators' awareness of environmental problems associated with the mining sector led to the adoption by the

Portuguese Government of a policy of accepting responsibility for remediating degraded and hazardous inactive mine sites, that for one or another reason have terminated their licences. Two preceding periods leading to the adoption of this policy can be identified.

In the first period, from 1995 on, the General Directorate for Environment (DGA) and the Mining and Geological Institute (IGM) agreed on the need for a general assessment of the environmental situation of former mines in Portugal. It commenced with the selection of three case studies representative of traditional mining activities. These were the Jales mine, an old sulphidic gold mine, Cunha-Baixa, a uranium mine where in situ leaching was used, and the Pejão coal (anthracite) mine. These were selected to develop relevant methodologies and to perform pilot studies within the scope of a protocol signed by the two institutions referred to above. Based on the results of these studies, namely on the results for the methodologies and technical solutions IGM, in cooperation with external consultants, in 1998 commissioned another set of studies on the environmental impact associated with the mines at Lousal-Caveira (pyrite ores), S. Domingos (copper ores) and Vale das Gatas (tungsten and tin). At the same time, using its in-house capabilities only, IGM undertook a preliminary assessment of a group of 85 old metal mines, excluding the uranium mines. This assessment led to a ranking of the sites into four different levels of environmental and sociological impact. At another level of detail, i.e. construction engineering, IGM awarded the engineering study and design project for the containment of the mill tailings dump at the Jales mine site. At the same time, the assessment and definition of priorities for remediating the group of radioactive ore mine sites was prepared by the National Uranium Company (ENU), the respective mining concessionaire [1, 2].

During the second period, from October 1999 on, the Ministries for Economics and Environmental Affairs agreed upon a cooperation protocol for environmental remediation and the implementation of legislative, organizational and financial measures in the mining sector. This protocol concerned the remediation of abandoned mine sites and the outline of policies in order to integrate environmental concerns into the practice of the current mining operations. This resulted in a cooperation agreement involving the IGM and DGA, as well as the State-owned Mining Development Corporation (EDM). EDM is a holding for the State's interests in the mining sector which, incidentally, had the most severe environmental problems associated with it, including the coal (Pejão mine), complex sulphides (Aljustrel mine), and uranium (60 sites with a very wide range of problems) sectors. The cooperation agreement aimed at developing a programme for the remediation of the abandoned and

contaminated mine sites, as well as the institutional and management framework needed for the implementation of the programme itself.

2. APPROVAL OF THE ENVIRONMENTAL REMEDIATION PROGRAMME

The solution finally adopted by the Government consisted of awarding an exclusive contract for implementation of the environmental remediation activities deemed necessary at the contaminated mine sites. The contract was awarded for a period of 10 years, is renewable, and comprises the characterization and assessment of the mine sites, implementation of environmental remediation works and the subsequent monitoring. Furthermore, the Government defined the conditions to be fulfilled by the contaminated mine sites in order to be included in the scope of the contract, which are:

- (a) Abandoned mining zones already decommissioned, whenever the former concession users cannot be made liable anymore, either as a result of bankruptcy or because the concession had been returned to the State earlier;
- (b) Sites at which mining activities ceased before the enforcement of the relevant mining regulation approved under Decree Law No. 90/1990, which contains provisions for the liability of mining companies for environment remediation;
- (c) Mining sites that still have a valid concession, but with a recognized need for an immediate intervention by the State for public safety reasons, as in the case of radioactive ores.

The main objectives of the remediation of contaminated mining areas are a sustained improvement of the sites in environmental, cultural and economic aspects through the elimination of risk factors that may represent a threat to public health and safety. These encompass risks such as soil and water contamination, exposure to toxic mining and milling residues. Relevant activities may involve remediation of the condition of the landscape and wildlife, preservation and improvement of the relevant archaeological heritage, as well as their recovery for economic and social use according to their specific suitability.

EXMIN — Industry and Mining Environmental Services S.A. (EXMIN), a services company entirely owned by EDM, was awarded an exclusive contract through Decree Law No. 198-A/2001 of July 6 2001 (the contractor). The contract was signed by the Government on September 6 of the same year, in the presence of the Ministers of Economy and Environment, and both

together were entrusted by the Council of Ministers to follow up on the implementation of the contract and related responsibilities on the programme's financing.

3. EMPOWERING THE STEERING COMMITTEE

The steering committee to represent State interests in the follow-up of this environmental remediation contract was also created by Decree Law No. 198-A/2001 of July 6, according to the terms of its appended Base XII. The members of the steering committee were nominated by Joint Notification No. 82/2002 and powers were bestowed on the committee by Joint Notification No. 83/2002, allowing the committee to start functioning as of 1 February 2002.

The steering committee is composed of five members, two being nominated by the Minister of Economics (one of them being the chairman of the committee), and one each by the ministers for environment affairs, of health and of science and technology (to whom nuclear and radiation protection matters are reported), respectively. The powers bestowed on the committee allow it to review all of the contractor's activities under the terms of the agreement, except the strategy of the remediation programme and its financing, which remain under the direct authority of the relevant ministers.

The steering committee is assisted by the technical evaluation subcommittee with competences for technical evaluation and follow-up of studies, technical projects and remediation field work undertaken by EXMIN. This technical evaluation subcommittee reports to the steering committee on the quality and adequacy of the solutions proposed by the contractor. The subcommittee is composed of a permanent group of three highly qualified technical staff representing departments of the Ministry of Environment — the former the institute for environment and later the institute of residues, the mining and geological institute, and the nuclear and technological institute, while three other members are nominated respectively by the regional authorities responsible for the environment, economics and health where the mine sites are located. When needed a representative of the archaeological institute is also on the subcommittee.

4. STRATEGIC AND MEDIUM TERM OBJECTIVES OF THE REMEDIATION PROGRAMME

With Joint Notification No. 83/2002, which formalized the bestowment of powers to the steering committee, the relevant ministers kept in their remit the

definition of the strategy to be adopted, including the selection of priority sites for intervention. It must be remembered that the mine sites are scattered throughout the whole of Portugal. They comprise more than 170 identified sites with preliminary characterizations already undertaken. This set of old mine sites encompasses a very wide heterogeneity of situations, such as the extent of the mine, type and pathways of contamination, environmental and social impacts, as well as the specific relevance of the threats to public health and safety. This already available information is a valuable basis for the comparative risk assessment study performed by EXMIN consultants. It is expected that this study, which is based on sound quantitative and qualitative parameters and optimized criteria, will result in the merging of the several different lists of mine sites requiring remediation into a single list with a clear definition of intervention priorities.

The basic data to feed such a comparative study come from studies performed by IGM and DGA between 1995 and 2001, as mentioned above. They include studies performed by the state owned Uranium National Company (ENU) and the contractor on a group of 59 former radioactive ore mines, and studies by the contractor on several complex sulphide ore mines, including Aljustrel (Pirites Alentejanas SA, PA). ENU is in liquidation, while PA is now dormant. These studies, plus the identification and assessment studies carried out by the contractor (EXMIN), allowed the number of former mine sites requiring some type of remediation to be fixed at 172.

The same contractor has prepared, from the year 2000 onwards, a set of relevant studies on the most urgent and relevant cases, namely sectorial comprehensive evaluations of both the complex sulphide ore and radioactive ore mines, case studies for Aljustrel and Urgeiriça, from which the relevant criteria for the general assessment and remediation plans for similar sites were derived. As a result, the engineering of the two most representative cases of the overall programme, Aljustrel, from the group of complex sulphides, and Urgeiriça, from the group of radioactive ores, respectively, reached an advanced stage. Detailed engineering designs that will allow initiation of field remediation work at these two sites will be under way in the near future.

The first level priorities already adopted took into account:

- (a) The importance of protecting public health and safety;
- (b) The need to distribute the activities throughout the country by adjusting and phasing the associated time schedules for the more complex cases and without putting the adopted overall solution at risk;
- (c) The time limit on the community support framework (QCA-III) to spend the funds allocated to the remediation programme;

- (d) The need to involve the local stakeholders, either public or private, to ensure the success of the programme.

Based on these priorities and other relevant conditions referred to above a preliminary definition of the respective allocation of funds for the period 2001 to 2006 was undertaken. The following spending is planned: 40% on the radioactive ore sites, 25% on the copper and pyrite ore sites, 20% on mine sites involving other base and precious metals (W, Sn, Au), while the remaining 15% are reserved for maintenance and monitoring and other technically and financially less important activities.

To finance the remediation programme from 2002 to 2006, a total of €56.2 million, with an average spending of €11 million each year, was allocated in 2001 to be released through the operating plans of the ministries for economics (€46.2 million under the economy operational plan — POE, later named PRIME) and environment affairs (€10 million under the environment operational plan — POA). Of the amount allocated to the programme about 20% must come directly from the State budget, as these are expenditures that are considered 'non eligible'. The remaining 80% are split between FEDER (75%) and the Portuguese State (25%), which means that in the end the real participation of the EU funds is limited to 60% of the total.

In the absence of any indication of future financial resources, the strategy the remediation programme adopted was to take a very long term perspective but to give priority to the most hazardous cases, which are generally also the more costly ones.

The programme will be concerned mainly with the remediation of mine sites abandoned by the former licence holder or already returned to the State. In two cases, however, the State assumed responsibility for remediation at mine sites while the licensee was still around:

- (1) Aljustrel mines, where part of the impact comes from ancient mining activities (namely during the Roman and African occupation of the country); part of the existing environmental responsibility was taken from the licensee, Pirites Alentejanas SA, and was subsequently assumed by the State; this occurred in 2000 as a result of negotiations with an international mining operator with a view to privatization of the company.
- (2) A group of 30 of the more than 60 radioactive ore mine sites under the concession of ENU SA; through Joint Notification No. 242/2002 of the Ministers of Economy and of Environmental Affairs; citing reasons of public benefit, the State assumed the environmental responsibility, subsequently integrating the group into the scope of the programme; in addition, as there is a plan to liquidate ENU in the near future, its

remaining radioactive ore mine sites will be subject to the same procedure.

5. REGULATORY AND SUPERVISORY FUNCTIONS OF THE STEERING COMMITTEE

The powers bestowed on the steering committee by the relevant ministers through Joint Notification No. 83/2002 can be summarized as follows:

“to approve the work programme and financial plans for a period of at least 3 years,...to approve the annual activity and financial budget, as well as the required adjustments,...to approve the remediation projects,... to obtain information from the Programme Contractor, ...as well as for other relevant matters.”

Such powers have been further detailed in Base XII, appended to Decree Law No. 198-A/2001, through the definition of the competences of the technical evaluation subcommittee, which include:

“to provide opinions on the quality and the adequacy of the solutions proposed,...and...to provide evaluations and execute technical reviews of studies, project engineering and site works, monitoring plans and reports, without exemption from the relevant mandatory studies and procedures on the environmental impacts...”.

Also, the obligations of the steering committee with respect to the project’s financing include:

“to issue an approval for each of the Contractors’ proposals calling for EU financing, concerning either preliminary and complementary studies or the subsequent remediation field works...”.

The steering committee, as a regulatory body, acted in a very proactive manner and, from the very beginning, defined a set of guidelines for the format and detail of the various types of project and financing documents to be submitted for approval by the contractor. These guidelines included, for example, a model for the conceptual master plan of each mine site and of the specific work plans and project proposals to be submitted to the relevant authorities in order to qualify for EU financing.

The committee considers the conceptual master plan for each mine site as being the most important item to be approved by them because it is the basic document to be used as a reference and for decision taking. These site specific master plans are intended to identify the existing situation, to outline an optimized technical solution for the problem, and collate the relevant objectives and conditioning parameters. Furthermore, they intend to:

- (a) Anticipate all possible consequences of the proposed intervention;
- (b) Explain the social context;
- (c) Illustrate economic and social benefits of the proposed remedial action at the site;
- (d) Investigate the relationship and participation of the landowner and other relevant stakeholders;
- (e) Anticipate the conditioning factors that may arise over the short or long term after the completion of work, such as monitoring and maintenance;
- (f) Evaluate the risk of future degradation of the improvements introduced and the cost of avoiding them;
- (g) Minimize the need for the long term interventions by the State, either technical or financial, working towards a sustainable solution.

Conversely, the investment proposals to be submitted to the relevant Portuguese authorities for obtaining EU financing, if based on the previously approved site master plan, have a more specific target, generally detailing the implementation of the proposed remedial actions. The main task of the steering committee is to verify that the proposed actions are clear, adequate, justified and in compliance with the relevant regulations. Nevertheless, the final decision on such proposals will be in the hands of the Portuguese management bodies concerned with EU financing, that is the POA, POE/PRIME and the ministers for environment affairs and economics, respectively.

6. THE CONTAMINATED RADIOACTIVE ORE MINE SITES

The environmental impact of the 59 radioactive ore mine sites is distributed over large areas of Beiras type granitic soil. The resulting environmental and public health detriments affect three districts and 15 municipalities. The Guarda district has 44 sites involving 10 municipalities, the Viseu district has 12 sites in four municipalities and the Coimbra district has three sites within one municipality. Of these municipalities, one has nine different mine sites within its boundaries, while four municipalities have seven mine sites each, one has six sites, three of them have three sites each, and the remaining municipalities contain one or two mine sites each. In addition, for 14 of the 59 mine sites there is a village or community immediately adjacent, and in five other cases at a distance of less than 500 meters.

The privately owned Companhia Portuguesa de Radium operated the uranium mines, which were permanently closed in 1999, between 1945 and 1962, whereupon they were transferred in 1977 to the state entity Junta de Energia Nuclear and later, in 1990, to the state owned company ENU. The

period of exploitation at each of mine site and the technologies used vary widely. Consequently, the levels of contamination also vary widely. The mine sites of Bica and Urgeiriça reached a maximum of 48 and 40 years of operation respectively, while at 16 mine sites operation lasted for only two years or less. The total amount of U_3O_8 concentrates produced during this period was in the order of 4400 tons, initially for stockpiling and later for export.

Aside from the environmental impacts originating from this period it is necessary to consider those that stem from an earlier period, between 1909 and 1945, when the production of radium was the exclusive objective of the operation. Mining operations during this initial period were not continuous, but fairly regular between 1911 and 1931, mainly at the Urgeiriça mine [3]. According to the available sources only about 50 g of radium seem to have been produced. In 1940 a progressive transition to the production of uranium concentrates was initiated.

The impacts from the mines operated during the 20th century are diverse, as they were caused by underground (29 sites), open pit (25 sites) or both types of mining together, exploited with different mining techniques. Mines have different problems associated with them, such as geotechnical instability including subsidence and instability of the ground or dumps of waste rock and middling dumps and of mill tailings ponds, residual contamination from underground and surface acid heap leaching, and drainage waters carrying heavy metals and other constituents detrimental to the environment. Impacts related to external radiation and radon emissions above permissible limits may vary from site to site.

The first strategic objective of the remediation programme is to hand back to the communities these contaminated and worked out mine areas in a condition that is acceptable from an environmental and public health point of view and where a minimum of restrictions on their final use will be imposed. Also, there should be minimal needs for monitoring and maintenance work. Nevertheless, it is anticipated that considerable care and a number of studies and consequent remediation actions will be required to achieve compliance with the relevant EU directives.

7. CURRENT STATUS OF THE PROGRAMME

By the middle of 2003 the containment of the mill tailings dump at the former gold mine at Jales had been completed, at a cost of €4.5 million as the first phase of its remediation plan; in 2004 a second phase involving complementary work for the treatment of residual acid mine waters was to have been initiated.

Following the completion of various preliminary assessment studies the conceptual master plans are being prepared for a few other sites in parallel with the respective environmental impact studies and the engineering projects. These will allow construction activities to start at some high priority sites. Work was to have been started in 2005 at three sites scattered around the country: at the Argozelo tin and wolfram mine (northeastern Portugal), supported by POA financing, and at the Uranium Urgeiriça (central zone) and pyrite Aljustrel (southern zone) mines, both supported with POE-PRIME financing.

Considering that the contract also allows the contractor to include in the scope of the programme the preparatory and preliminary studies executed before the contract was awarded, the total expenditure by the programme by the end of 2003 amounted to around €12 million. Considering that the EU funds available for this specific activity are to be spent by the end of 2006, the work plans need to be adjusted so that the corresponding disbursement reaches a level of €15 million per year, which is considerably higher than the average performance over the past three years and now seems to be an extremely difficult target to achieve.

Although the former government policy of implementing the remediation programme is strongly maintained, the perspectives for the year 2004 were not optimistic, taking into account the country's present economic situation, aggravated by the difficulties due to the reduction of the State Budget deficit to comply with the limits imposed by the EU, which will result in substantial restrictions on the availability of funds required to cover the nearly 40% of the total costs to be borne by the Portuguese State.

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Paper 21

GEOCHEMICAL EVALUATION FOR THE SITE CHARACTERIZATION OF THE CUNHA BAIXA URANIUM MINE IN CENTRAL PORTUGAL

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Abstract

The Portuguese Mining and Geological Institute carried out a number of studies which broadly aimed at investigating the environmental impacts of abandoned mines in Portugal. These studies served to obtain information for site characterization prior to further remediation steps. The Cunha Baixa mine, located in central Portugal in Mangualde county, is reported as one important case study. A geochemical survey involving analysis of stream sediment, alluvial soil, rock and tailings samples collected around the Cunha Baixa mine was carried out in order to delineate the geochemical signature and detect the pollution focus. Some groundwaters were also collected in wells and boreholes. From the results obtained we can say that remediation is necessary to protect environmental and public health in Cunha Baixa.

1. GEOLOGY, MINE HISTORY AND WATER TREATMENT

The Cunha Baixa mine is located in central Portugal, in Mangualde county [1–4]. The ore deposit comprises brecciated quartz veins that fill two main N40°E and N70°W striking fractures (Fig. 1) [5]. Uranium was exploited in the Cunha Baixa mine for about 30 years. During the period 1967–1983 underground stoping methods were used, but later (between 1984 and 1991) in situ chemical leaching procedures using sulphuric acid were adopted. In total, some 484 000 t of ore with an average grade of 0.186% U_3O_8 were produced. The mine produced more than one million tons of sterile waste and tailings materials, which were partially used to fill a 300 m × 100 m open pit (located immediately above the underground works of the Cunha Baixa mine) and also deposited nearby as coarse grained tailings [6]. About 1 km to the west of the Cunha Baixa mine another open pit operated into the recent past (the Quinta

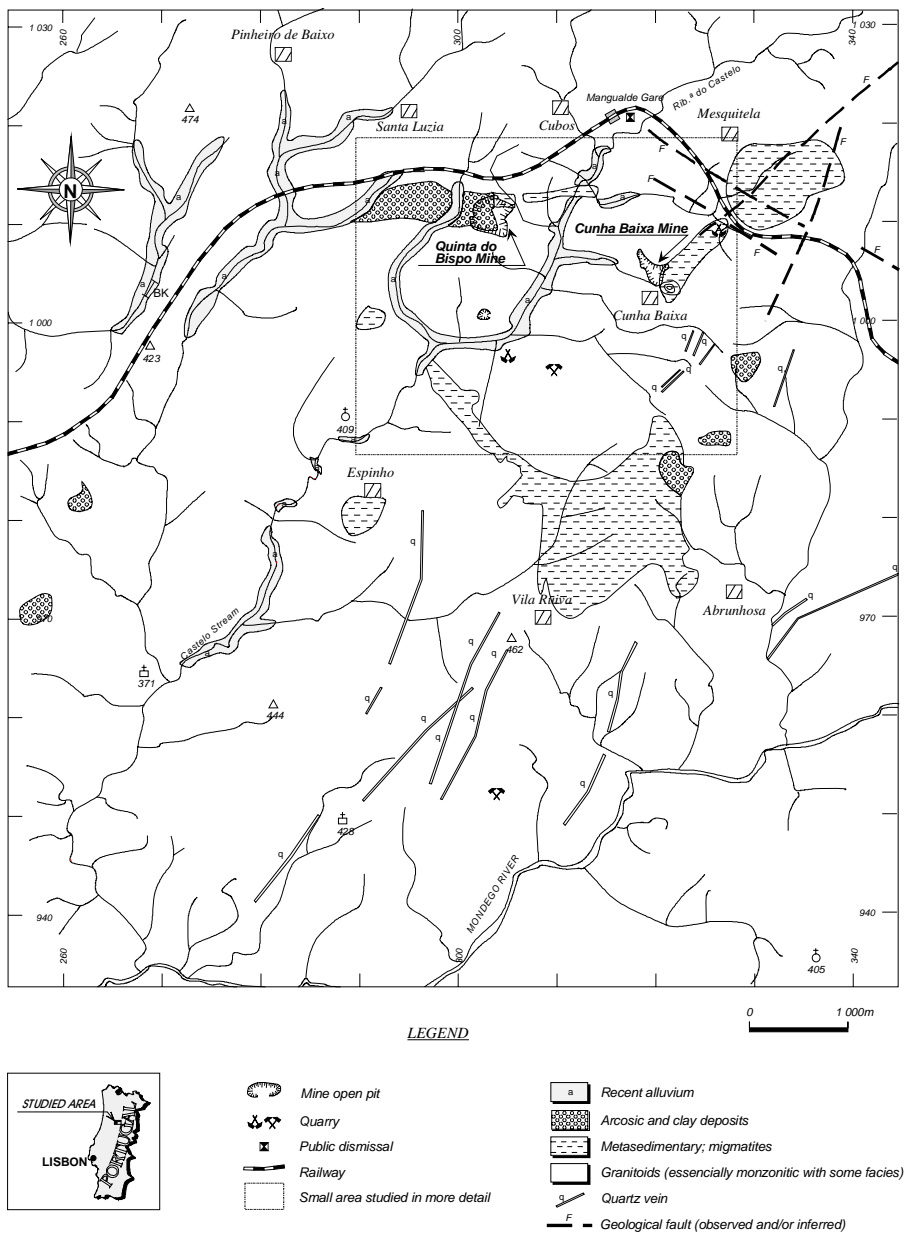


FIG. 1. The Cunha Baixa area: Study area and geological setting.

do Bispo mine). Low grade ores there were treated using similar leaching procedures.

The water remaining inside the Cunha Baixa mine is pumped continuously to the surface through the main shaft, where it is chemically neutralized with calcium hydroxide. Despite this, some chemical contamination is found in surface water draining from the mine, as well as in some groundwater. Barium chloride is added to precipitate minor quantities of radium in the water. The slurries formed by these operations, mainly composed of gypsum, are deposited in an old open pit nearby. Groundwater is used by the local population mainly for irrigation and (rarely) as drinking water, while soils in the vicinity of the two mines are used for agricultural purposes. Contamination of these media by mine waste contamination is the principal concern of the local population.

2. GEOCHEMICAL ANALYSES

Multi-element analyses (DC plasma spectrometry and FRX complemented with fluorimetry for uranium) were carried out on a number of stream sediment, alluvial soil, rock and tailings samples collected around the Cunha Baixa mine. Trace element composition of the mine tailings strongly contrasts with those of the geological basement (mainly granitic rocks). This is particularly evident for uranium and to some extent for zinc and copper (Table 1), reflecting the influence of the mineral paragenesis (uranium minerals and some sulphides) that appears to still be imprinted in the tailings. This strong U enrichment (with an average of 318 ppm in the tailings material) also defines an anomalous level when compared with some established baseline values for U, such as 2.7 ppm in the Earth's crust [7], 3.9 ppm in granites [8], and 9.5 ppm in granites of central Portugal [9].

Uranium turned out to be the only element geochemically anomalous in stream sediments (Table 2). High concentrations were detected, either for the total uranium (U_t), or for the more mobile phases, i.e. the leached uranium (U_l). Among the other hazardous metals, only Zn, Cu and Mn show subtle anomalies in places. Statistically, U seems to be independent of the majority of the other analysed elements.

As would be expected from ^{238}U isotope decay [10], ^{226}Ra was present. Analyses carried out in alluvium, stream sediment and tailings samples gave average values of 444, 1313 and 2647 Bq/kg respectively, demonstrating the existence of radioactivity in areas closest to the mines. Alluvial soils collected near and downstream from the mine sites are slightly contaminated by uranium (Table 3). However, lower concentrations were found in the soils than in stream sediments (an average of 30 ppm of U versus 478 ppm in downstream

TABLE 1. AVERAGE CONTENT OF U AND OTHER SELECTED CHEMICAL ELEMENTS IN THE TAILINGS AND ROCKS OF THE AREA OF THE CUNHA BAIXA MINE

Sample type	U _t (ppm)	U _l (ppm)	Cu (ppm)	Zn (ppm)	As (ppm)	Ni (ppm)	Co (ppm)	V (ppm)	Cr (ppm)	F (ppm)	Fe %	Mn (ppm)	P (ppm)
Tailings (n = 13)	318	161	41	120	<20	18	13	18	186	840	2.4	367	1000
Granite (n = 4)	9.3	7.4	8	66	<20	8	<10	7	170	1192	-	394	-

TABLE 2. AVERAGE CONTENT OF U AND OTHER SELECTED CHEMICAL ELEMENTS IN STREAM SEDIMENTS OF THE AREA OF THE CUNHA BAIXA MINE

Sample type	U _t (ppm)	U _i (ppm)	Cu (ppm)	Zn (ppm)	As (ppm)	Ni (ppm)	Co (ppm)	V (ppm)	Cr (ppm)	F (ppm)	Fe %	Mn (ppm)	P (ppm)
(1) Overall data	M	118	75	67	130	20	14	10	11	900	2.2	689	1131
(n = 61) using samples collected at random in the area (see Fig. 1)	sd	262	139	55	88	15.2	15.2	11.2	10.4	250.4	0.6	638	406
(2) Values referred to anomalous samples closed to the mine sites (n = 11)	M	478	235	77	224	26	19	22	26	895	2.7	912	1658
	sd	461	217	41	140	19.6	15	17	26	268.6	0.7	591	498
(3) Values referred to geochemically barren samples (n = 11)	M	14	10	32	86	11	6.4	5.5	7.1	630	1.9	430	994
	sd	2.6	2.7	16.1	12.7	3.9	2.3	1.5	2.9	176.9	0.4	173	248
(4) Reference statistical parameter for the local background, data from (3)	M + 2 × sd	19	15	64	111	19	11	8	13	984	2.7	776	1490

M = Arithmetic mean; sd = Standard deviation

TABLE 3. AVERAGE CONTENTS OF U AND OTHER SELECTED TRACE ELEMENTS IN ALLUVIAL SOILS AROUND THE CUNHA BAIXA MINE

Samples	U _t (ppm)	U _i (ppm)	Cu (ppm)	Zn (ppm)	As (ppm)	Ni (ppm)	Co (ppm)	V (ppm)	Cr (ppm)	Fe %	Mn (ppm)	P (ppm)
Alluvial samples close to the mine workings (n = 55)	M 30	18	42	93	12	5	5	11	57	2	432	935
	Sd 33.7	11	24.4	15.4	6.3	—	1.2	3.4	15.7	0.3	83.5	249.6
Samples corresponding to local background	M 12	10	47	93	~10	~5	~5	8	62	1.8	381	1098
	Sd 2.2	2.1	24.9	26.7	—	—	—	3.6	8.6	0.3	91.4	462.2

M = Arithmetic mean; sd = Standard deviation

sediments, probably due to the high solubility of uranium over a wide pH range) [11]. According to Neves et al. [12] these soils are acidic with weak ion exchange capacity that is due to low organic matter content. It is thought that intense agricultural use of these alluvial soils with frequent use of phosphate fertilizers can also be contributing to uranium leaching and later accumulations in other places.

The effects of contamination appear to be more strongly imprinted in the neighbouring stream sediments. The spatial distribution of U along the main stream (and its tributaries) directly influenced by the two mines shows persistent chemical anomalies that occur at least 10 km away (Fig. 2). However, these anomalous patterns are irregular along the main stream, suggesting the existence of superimposed chemical and mechanical processes in the secondary uranium distribution. This kind of secondary dispersion process for uranium has been found by other researchers [13]. The investigation of the Cunha Baixa mine is important in demonstrating the occurrence of uranium in soils and water as an industrial contaminant.

3. HYDROCHEMISTRY

Some groundwater samples collected in wells and boreholes confirm the existence of strong anomalies for sulphate, Ca and other metals (Table 4),

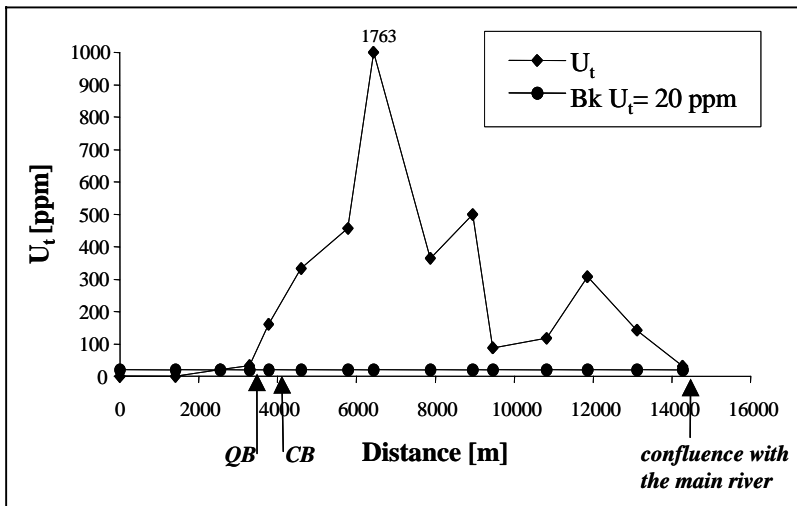


FIG. 2. Dispersion of U in the sediments of the main stream draining from the mining sites (CB: Cunha Baixa mine influence; QB: Quinta do Bispo mine influence).

TABLE 4. AVERAGE VALUES OF SELECTED HYDROCHEMICAL PARAMETERS IN GROUNDWATER AROUND THE CUNHA BAIXA MINE

Sample location	SO ₄ ²⁻ pH (ppm)		Mn (ppb)	Zn (ppb)	Co (ppb)	Ni (ppb)	Al (ppb)	Be (ppb)	Y (ppb)	Sr (ppb)	U (ppb)
About 1 km NW of the Quinta Bispo mine	6.2	6.2	85	~9	<9	<29	<50	<5	<4	37	3.8
Up to 1 km SE of the Cunha Baixa mine	6.0	17	29	93	42	<29	<50	<5	<4	66	21
Mine influenced samples	5.1	829	6096	1006	63	333	4680	91	337	1085	912

including significant concentrations of U together with those of Mn, Zn, Co, Ni, Al, Be, Y and Sr. Concentrations in the range of hundreds of ppb (ppm for sulphate) are common for most of the elements, in particular for sampling locations more or less between the two sites. Samples taken at greater distances (roughly between 0.5 and 1 km to the SE and NW, respectively) show less contamination and, in general, show low contrasting concentrations for the majority of the elements.

The chemical data suggest that mine contamination has occurred due to infiltration into the groundwater resulting from the ore leaching process. In addition, some of the contamination probably resulted from uranium and other metals that were released from the residual slurry and subsequently released to the water. On the other hand, surface waters collected downstream from the drainage system do not reveal such critical patterns. Conversely, they seem to show a very fast decay pattern and a very common geochemical signature of $\text{Al} + \text{Mn} + \text{Zn} + \text{SO}_4^{2-}$, apparently always present in surface waters that come from mine sites (Fig. 3). In this context the Cunha Baixa is environmentally similar to other case studies with a very different paragenesis.

4. HYDROGEOLOGY

Granite is the main rock type of the underlying geology. These massifs are strongly fractured (faults, small fractures, fissures, diaclases) due to tectonic influence. The structural features, which are particularly significant around the

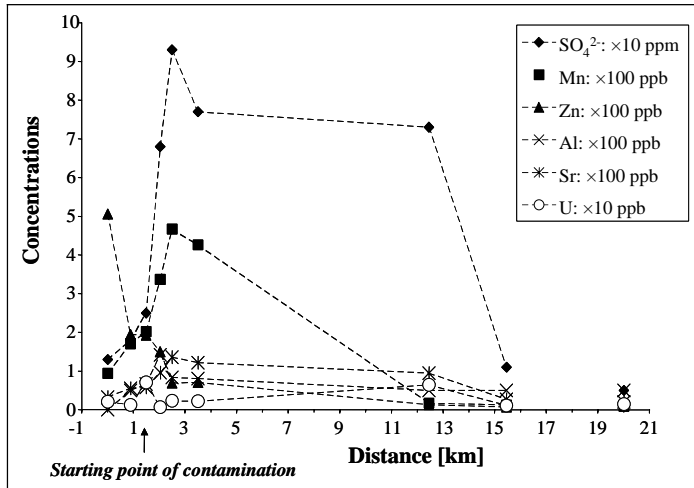


FIG. 3. Dispersion of U, other metals and sulphates in surface waters of the main stream draining from the mining sites.

deposits, greatly increase the permeability of the rocks and their hydraulic behaviour. A preliminary hydrogeological study showed that water wells (deeper aquifers) closer to the mine sites are strongly contaminated. They appear to be located along selected fault strikes, suggesting that some of these systems constitute preferential water conduits. Moderate enrichments in some metals were detected (Zn, Co and U) in many of the boreholes, some of which can be detected up to about one hundred metres in depth. Beyond these geological structural controls, modifications in redox and pH conditions, interactions between phases (rock/water) and the microbial degradation of organic matter can also contribute to metal dispersion and accumulation in groundwaters [14].

On the other hand the wells that tap shallower aquifers are in general placed in more porous rocks (altered granites, sedimentary deposits). Owing to this, they show a stronger enrichment in sulphate and in most of the metals analysed (Mn, Zn, Co, Ni, Al, Y, Sr and U), together with lower pH values. These data are summarized in Table 5.

Figure 3 summarizes the main uranium data obtained for the area surrounding the two uranium mine sites. The combined interpretation of the element distribution in stream sediments, alluvial soils, rocks, waters and tailings can assist in building a geochemical–hydrochemical–hydrogeological model of the uranium in the study area, which is greatly influenced by the presence of ‘mining signatures’.

TABLE 5. AVERAGE VALUES OF SELECTED HYDROCHEMICAL PARAMETERS IN WATER BOREHOLES AND WELLS UNDER THE INFLUENCE OF THE CUNHA BAIXA MINE

Samples	pH	SO ₄ ²⁻ (ppm)	Mn (ppb)	Zn (ppb)	Co (ppb)	Ni (ppb)	Al (ppb)	Be (ppb)	Y (ppb)	Sr (ppb)	U (ppb)
Boreholes	6.4	4.5	65	334	11	<29	63	5	<4	27	190
Wells	4.7	1065	7819	1199	49	424	5998	116	433	1388	1118

5. CONCLUSIONS

The necessity for remediation to protect the environment and public health has been demonstrated by this multidisciplinary study. Compounding this problem, about one million tonnes of tailings with an average residual content of 320 ppm U (accompanied by some ²²⁶Ra) still exist in the Cunha Baixa open pit.

Efforts undertaken by the mining company (ENU until recently) to control and mitigate possible impacts from their previous extraction operations were successful to some degree. However, the toxic character of uranium, radium, as well as other chemical elements with significant biological activity [15–17] (namely Be, Sr and Y), together with significant concentrations of a variety of other harmful elements in waters (Al, Mn, Zn, Ni) that have been identified, are a matter of concern. Also, some alluvial areas adjacent to and downstream from the mine sites with ‘uraniferous’ contamination will place constraints on future land use.

Scientific knowledge of the studied area is still limited. To overcome this, proposals were initiated to more effectively characterize the nature and extent of contamination:

- Detailed hydrogeological studies of the mine drainage basin;
- Mineralogical and geochemical research (detailed identification of the existing U minerals; partition of the uranium within the different mineralogical phases using chemical techniques of metal selective extraction);
- Toxicological investigations on the degree of hazard due to the most harmful elements;
- Epidemiological studies to investigate the degree and extent of certain human diseases in the area and possible correlation with toxic elements;
- Monitoring for water quality, giving particular emphasis to ²²⁶Ra.

Further to these studies, effective and sustained measures aimed at promoting remediation of the mine sites and surrounding contaminated areas must be developed.

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Paper 22

RADIOLOGICAL CHANGES DUE TO THE RESTORATION OF A URANIUM MINE

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Abstract

The paper presents a radiological study of a uranium mine in Extremadura, a region in the southwest of Spain. Mining work had already ceased and coincident with the development of this study the mine was being remediated, making it possible to study the influence of the remediation plan on different radiological parameters. The radiological parameters were studied in three groups: aerial samples, soil samples, and water sediment samples. The first two groups were clearly influenced by the remediation work, but the third was not. The results for ^{222}Rn in air and water, ^{222}Rn exhalation, effective ^{226}Ra in soils and sediments, natural uranium and ^{226}Ra in water, and the physico-chemical and meteorological parameters, allowed us to characterize the zone of the mine. Particular variations in certain radiological variables are explained by taking into consideration some of the physico-chemical parameters measured.

1. INTRODUCTION

The importance of the radiological impact of uranium mining and milling has long been recognized. Evaluation of this impact is therefore of general concern, but to be meaningful it must be followed along each phase of mining

activity. Also, even though mining and milling have ended, an impact on the environment is still likely and hence remediation work becomes necessary to reduce the effect of the radiological impact of the installation to background levels. In view of this situation, we were prompted to carry out a study on the effects of a remediation programme that was implemented in the area of a uranium mine in southwestern Spain. Several campaigns coincided with different steps in the remediation programme, such that it was possible to study its effects on several radiological parameters: ^{222}Rn in air samples, ^{222}Rn exhalation, effective ^{226}Ra in soil samples, and ^{222}Rn , ^{226}Ra and natural uranium in ground and surface water. Several physico-chemical parameters were also determined in order to discriminate the effects due to the remediation from those due to other, natural processes.

2. MATERIALS AND METHODS

2.1. Study area and sampling sites

This study was performed from 1996 to 1998 (coinciding with the remediation programme) at the 'El Pedregal' mine, the exploitation of which ceased in 1990. This mine is located in Extremadura, a region in the southwest of Spain, about 8 km from the village of La Haba. Different types of farming and agricultural activities are being carried out in its surroundings. Two small rivers, tributaries of the River Ortigas, called the Madroñal and the Pilones, bordered the mine (Fig. 1). In addition, there are several sources of groundwaters in the zone, some of them occasionally used for drinking.

In Fig. 1, sampling points are identified as follows: inside the mine's fence, ES groups the three most important slag heaps of the mine; point CO indicates the original open pit which had already been remediated before the time of this study; LD is the slushpit of the mine, with a release of water towards a small reservoir represented as DL; DI represents the tailings dam of the mine. Points OA, MA and PA belong to the rivers Ortigas, Madroñal and Pilones, respectively, upstream from the installation. OB, MB and PB represent points of the same rivers downstream and hence potentially affected by the mine. SP, SS and PO are sources of underground water, located inside or near the boundaries of the installation. Finally, other reference points outside the installation are HA (in La Haba, to the North, off the map), CA, ZA and CD.

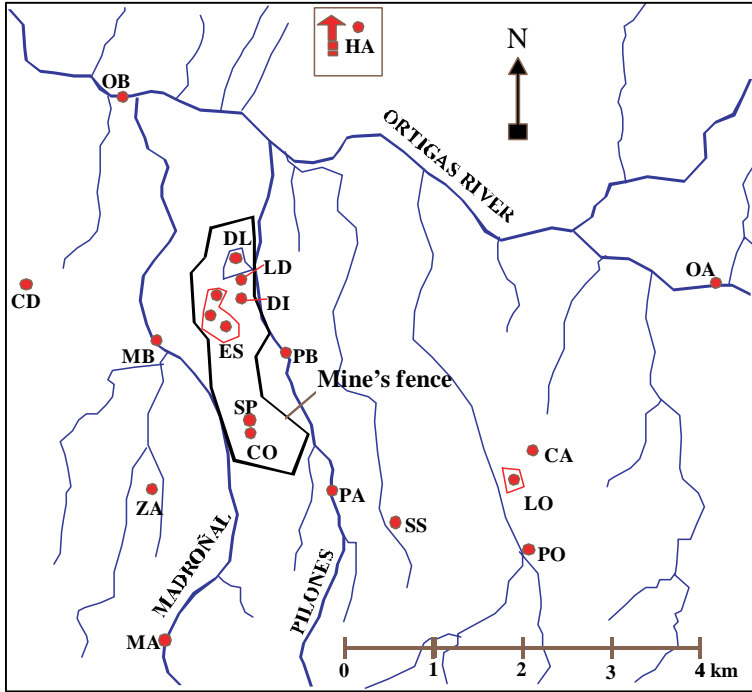


FIG. 1. Map of the mine. The sampling sites are marked.

2.2. Sampling plan and measurement techniques

Three sampling campaigns were carried out. The first and third were quite similar as regards hydrological and meteorological conditions, corresponding to wet and cool weather and moderate hydrological conditions. The second was characterized by dry and warm weather and static hydrological regimes.

Air samples were collected to determine ^{222}Rn activity concentrations. The reference points were all outside the mine (HA, OA, CA, MA, ZA, CD, OB), while sampling was carried out at points ES, LD, CO and DI inside the fence. ^{222}Rn was sampled using a dynamic device and measured by liquid scintillation counting (LSC) [1].

^{222}Rn exhalation and effective ^{226}Ra in soil samples were measured at the same points. Only point (ZA) was considered as a reference. Another two points (CO, ES) were not affected by the restoration work under way during this study, while a further two points (DI, LD) were selected to evaluate the

local effects of the restoration. Sampling and measurement procedures can be found in Ref. [2].

Water samples of underground origin (SS, SP and PO) were collected for ^{222}Rn , ^{226}Ra and uranium measurements. The same analyses were performed for the samples of surface origin: OA, OB, MA, MB, PA, PB. Specific procedures were used for ^{222}Rn , ^{226}Ra [3] and natural uranium [4] activity determinations by LSC.

3. RESULTS AND CONCLUSIONS

Following are some results for the radiological variables considered. They are presented as averages of several points, grouping the values corresponding to each campaign and, in specific cases, highlighting their significance with regard to certain advances in the restoration programme.

Table 1 shows the results for ^{222}Rn in air for the three campaigns. The sampling points are grouped in reference (outside the mine) and internal points. As can be seen, the highest values were found inside the installation, mainly during the first campaign (the highest value was $38 \pm 4 \text{ Bq/m}^3$ in the LD point). According to the results of the different campaigns, a clear decrease occurred in the values of ^{222}Rn in air inside the mining area, which can be associated with the remediation programme. In the third campaign, the results obtained outside and inside were statistically indistinguishable, with an overall average value of 2.1 Bq/m^3 . This result is consistent with reported values of ^{222}Rn in air for zones with no direct influences [5].

For ^{222}Rn exhalation, in the first campaign an average value of $3509 \text{ Bq/m}^2/\text{h}$ was obtained, ranging from 429 to $10\,507 \text{ Bq/m}^2/\text{h}$, while in the second and third campaigns the average value was strikingly reduced to 831 and $336 \text{ Bq/m}^2/\text{h}$, respectively. Thus, as expected, the remediation work is clearly reflected in the values of this radiological parameter. Maximum values were all found in the first campaign: $10\,507 \pm 237$ and $4\,557 \pm 117 \text{ Bq/m}^2/\text{h}$ at points DI and LD, respectively. The average value for the first campaign clearly surpassed the $2664 \text{ Bq/m}^2/\text{h}$ threshold considered for remediation work as required by, e.g. the US Environmental Protection Agency [6]. After completion of the remediation programme, in the third campaign no sampling points gave values above this threshold (the maximum value was $750 \text{ Bq/m}^2/\text{h}$ at point CO).

Effective ^{226}Ra in soil was determined in samples collected at the same points as those used to test for ^{222}Rn exhalation, both showing very similar behaviour, i.e. a clear decrease in the concentration due to the remediation programme (from an average value of 2170 Bq/kg in the first campaign to 581 Bq/kg in the third campaign).

TABLE 1. AVERAGE VALUES (RANGE) OF ^{222}Rn ACTIVITY CONCENTRATIONS IN AIR (Bq/m^3)

Campaign	Outside	Inside
1	4.6 (3.9–5.8)	16.5 (6.1–38)
2	3.3 (2.3–4.8)	5.3 (2.3–12)
3	2.1 (0.79–2.9)	2.2 (2.0–2.3)

As expected, ^{222}Rn activity concentrations in groundwater were considerably higher than for surface waters. The average value for groundwater (SS, SP and PO) for all the campaigns was 282 Bq/L (range between 22 and 669 Bq/L), unlike the averages of 2.0 Bq/L (range, 0.86–3.2 Bq/L) for surface waters upstream (OA, MA and PA), and 5.7 Bq/L (range, 0.30–12 Bq/L) for points downstream (OB, MB and PB). In order to evaluate the possible effects of the remediation work on ^{222}Rn activity concentrations in surface waters, the data for points upstream were compared with the corresponding points downstream for each campaign using the student's t-test. For the first campaign, the results showed that downstream waters have higher activity concentrations than points upstream (at the 90% confidence level), although both data series were already indistinguishable (95% confidence level) by the third campaign. This suggests slightly reduced effects due to the remediation work.

A high degree of variability was found for ^{226}Ra activity concentrations in surface waters: for example, at point DL the values obtained were: 0.10 ± 0.03 , 0.026 ± 0.006 and 2.0 ± 0.5 Bq/L in the first, second and third campaigns, respectively. The observed trend does not seem to reflect any effect of the remediation programme. Indeed, an important increase was observed in the third campaign. This trend cannot be explained in terms of possible concentration dilution effects (the last campaign was the wettest). Instead, an explanation can be found in the sulphate concentration in the water, which followed an inverse trend in each campaign: 33.4, 86.2 and 13.5 mg/L, respectively. This behaviour can be interpreted to mean that a higher concentration of sulphate in the water leads to precipitation of insoluble sulphate salts, which then co-precipitate dissolved ^{226}Ra .

Finally, with regard to natural uranium in water, the variations in the values can be better explained in terms of dilution effects (hydrological conditions) and the presence of dissolved carbonates (or pH), than as being due to the remediation work. In general, the highest values were recorded in the second campaign, in good agreement with the concentration effects in the waters. The highest uranium concentration was always found at DL, which can

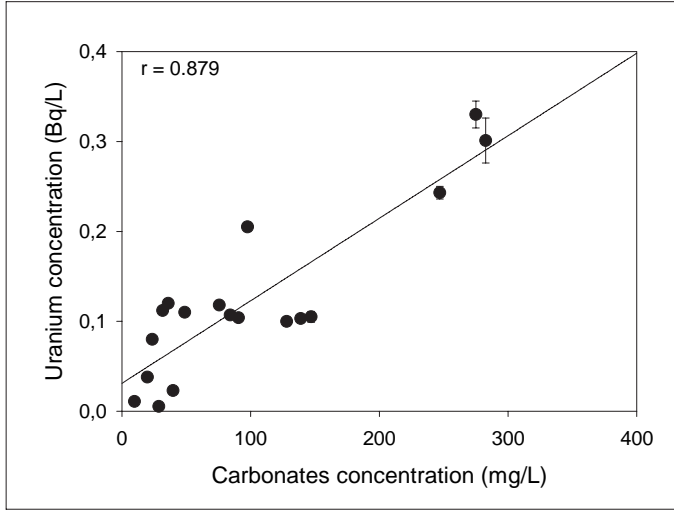


FIG. 2. Variation in the natural uranium concentration with the concentration of carbonates.

be explained in terms of the special features of this point. On the one hand, this point collected all the runoff waters from the mine; on the other, this point had certain chemical particularities, especially its very low pH (3.5–4.8). Except for DL, where low pH values explain the lowest presence of carbonate, a close correspondence between carbonate concentrations and uranium activities was observed at all points (Fig. 2).

In a broad context, and during the period of this study, it seems that the radioactive impact of these mines was not very large off-site: the activity concentrations of ^{222}Rn in air and ^{226}Ra and uranium in water, which show the highest mobility, do not appear to be significantly elevated outside the mine's limits compared to the levels observed at other locations not affected by the presence of this type of installation.

On the other hand, the remediation work has had a major effect on soils (^{222}Rn exhalation and effective ^{226}Ra) and air (^{222}Rn) inside the mine's fence, reducing their activities down to levels typical of the zone. The remediation work did not produce any remarkable effect on the concentrations in waters, the variations in activity of which can largely be explained in terms of meteorological variables (such as that the concentration effect is increasing the activity observed in dry conditions) or other physico-chemical variables (pH, sulphate or carbonate concentrations).

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The legacies of past uranium mining and milling activities continue to be of concern and require assessment and remedial action. In some countries there are numerous small-scale mines and mills. For economic and other reasons, including less stringent environmental standards and awareness at the time, these operations may not have been properly closed out and made safe. An international workshop was organized in Lisbon from 11 to 13 February 2004 as a forum for the exchange of views and experiences of countries with smaller scale uranium mining legacies. These proceedings feature a summary of the workshop, including the major conclusions, as well as the technical papers presented at the meeting.