

IAEA-TECDOC-370

**ENVIRONMENTAL MIGRATION
OF RADIUM AND OTHER CONTAMINANTS
PRESENT IN LIQUID AND SOLID WASTES
FROM THE
MINING AND MILLING OF URANIUM**

**FINAL REPORT
OF A CO-ORDINATED RESEARCH PROGRAMME
SPONSORED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
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FOREWORD

This document is the final report of the Co-ordinated Research Programme (CRP) on "The Environmental Migration of Radium and Other Contaminants Present in Liquid and Solid Wastes from the Mining and Milling of Uranium. Its main content is a discussion, with recommendations, on the outstanding needs for research in this subject area. The main purpose of the CRP was to facilitate the exchange of information between interested scientists in Member States of the IAEA. The participants have shared many discussions on the current state of knowledge in the subject area and the discussion section is a reflection of their deliberations. In addition, the document contains a considerable amount of useful reference material concerning details of participating scientists, their relevant publications and their projects within the CRP.

It has also to be mentioned that this CRP following, as it did, a previous CRP on a related subject* has led to an Agency project, now underway, to produce a Monograph or textbook on "The Environmental Behaviour of Radium". The Monograph will contain contributions from over 50 experts in the field and will represent a unique review of information in this subject area. The project is being guided by a Steering Committee made up of several of the participants of this and the previous CRP.

The CRP was organized on behalf of the Agency by Jacques Molinari who was Scientific Secretary from its inception in 1981 to 1984. Gordon Linsley was Scientific Secretary for the final Research Co-ordination Meeting in 1985.

* CRP on "The Source, Distribution, Movement and Deposition of Radium in Inland Waterways and Aquifers" (1976-1980)

EDITORIAL NOTE

In preparing this material for the press, staff of the International Atomic Energy Agency have mounted and paginated the original manuscripts and given some attention to presentation.

The views expressed do not necessarily reflect those of the governments of the Member States or organizations under whose auspices the manuscripts were produced.

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1. INTRODUCTION*

1.1. Background on Uranium Mining and Milling Wastes

Most of the public health risks and environmental impacts associated with uranium mining and milling are common with those of the base metal mining and milling industry. However, uranium mining and milling presents the possibility of additional public health effects which arise because of the greater amount of radioactivity in the ore.

At the end of mining and milling, waste materials that could harm the public and the environment are left at the mine and mill site. Deciding what is the best method for the management of these wastes is difficult and the decisions made for one site may be quite different from those for others.

The wastes from actual mining operations that have to be dealt with in the long term are waste rock and ore containing concentrations of uranium that are uneconomic to process. These wastes may pose environmental and public health problems, especially if they contain a substantial level of pyrite (FeS_2) and heavy metals. The nature of these wastes depends on ore and host rock mineralogy; the quantity depends on the orebody configuration and mining methods.

The principal wastes from milling uranium ore are the tailings; the sands and slimes which are the finely ground residues from ore processing. They are produced in large quantities, their mass being close to that of the ore which was processed.

Many non-radioactive contaminants (e.g. heavy metals) may have undesirable impacts on some flora and fauna, but, with some notable exceptions (e.g. As, Hg, Cd, Se), rarely pose a direct health risk to man. In contrast, many of the radioactive daughters of uranium, while of little direct consequence to the environment, pose a radiological health risk to man. In most cases the risks to individuals are small compared with other risks routinely experienced. However, for both non-radioactive contaminants and radioactive contaminants (because of the long half-lives of the parents) these potential impacts continue into the distant future. The behaviour of the wastes have to be considered over times that are unusually long compared with human experience.

1.2. Radionuclide Releases

Radon-222, an inert radioactive gas with a 3.8 day half-life, continually escapes to the atmosphere from radium-226 in all surface soils. The escape rate from uranium mill tailings is usually higher than from normal soils depending on the physical properties of the tailings and any cover over the tailings. The distribution of Ra-226 may vary with time. Leaching may result, for example, in a downward migration of Ra-226 from the surface - or the effective source area may increase if the tailings disperse through erosion. The properties of any cover may change; for example, the thickness or integrity may be reduced by erosion.

* Much of the introductory material in this chapter is extracted from "Long-term radiological aspects of management of wastes from uranium mining and milling", OECD/Nuclear Energy Agency, Paris (1984).

The radiological risk to the public from Rn-222 results from the irradiation of lungs by the short-lived airborne alpha-emitting daughters of Rn-222. At and very close to the tailings the concentrations of these nuclides in air can be measurably greater than what are regarded as normal background levels. The highest exposure to Rn-222 released from tailings might be of individuals who build houses and live on actual tailings sites. At the other extreme, if the tailings are water-covered or are covered by deep snow, then any exposures to radon will be very much reduced.

Beyond a few kilometres from a tailings area any increase in radon concentration owing to releases from that area cannot be distinguished from background levels. Thus radon released from tailings can produce an immeasurably small but nevertheless calculable increase in concentrations of radon in the atmosphere distant from the tailings. Since it only takes two or three weeks (3 to 4 half-lives of radon-222) for weather patterns to circumnavigate the earth such increases are calculable on a global scale. Thus, very small fractional increases (above natural background levels) in radiation doses to populations from radon daughters can be estimated to result from uranium mill tailings.

Radionuclides such as Th-230, Ra-226, Pb-210 and Po-210 may be leached from waste or tailings piles. The leach rates depend on the hydrogeochemistry of the tailings and surrounding rock or unconsolidated sediments, on the integrity of any retaining structure and on the permeability of any capping material. These factors are likely to change with time. People may be exposed to radiation from the leached radionuclides as a result of transfer along aquatic foodchains and through contamination of drinking water. Aquatic exposure pathways are particularly important in wet climates. Close to a tailings site the concentration of these radionuclides may be sufficiently high for the contribution from that site to be measurable but further away such contributions to the normal levels in diet and the environment are only calculable.

Radionuclides in waste rock particles or tailings dust may be dispersed by wind and water erosion if the waste piles are not contained by dams and cappings, or if such containment has been breached as a result of natural erosion or human intrusion.

The erosion rates of cap materials for tailings will depend on the engineering design of the capping and on geomorphological processes. Values can be estimated for these rates under current conditions. Rather more speculative are estimates of the erosion rates of tailings that become uncovered after having undergone hundreds or thousands of years of internal geochemical activity. The pathways to humans could be by inhalation, by transport through aquatic foodchains, or by deposition and transport through terrestrial foodchains. Direct external irradiation from dispersed material may also occur.

Tailings could also be removed by humans for misuse such as for building materials or for land fill. External irradiation or exposures to Rn-222 and daughters at levels unacceptable to the public can result from such practices.

1.3. Radiological Impact

The pathways leading to the greatest exposure of individuals in the population vary depending upon the conditions at the site. For example, aquatic pathways may vary from being dominant in importance as contributors to individual dose in wet climatic conditions to being

insignificant in dry arid regions. Annual effective dose equivalents have been estimated to range between a few hundred uSv to several mSv for individual members of the public living near to typical mines and mills (UNSCEAR 1982).

Although it is clear from the foregoing discussions that the radiation exposure of individuals, even those living fairly close to the facilities, will be very small, because of the extended dispersion of radon and its daughter products in the atmosphere their collective radiological impact will be significant. The collective effective dose equivalent commitments from mining and milling wastes have been estimated to contribute the major part of the collective radiological impact of the nuclear fuel cycle even though more than 90 per cent of the collective dose is made up of individual annual effective dose equivalents of less than 1 uSv (that is $\sim 0.1\%$ of that due to natural background radiation (typical)) (UNSCEAR 1982). Because of the widespread dispersion of radon (albeit at very low concentrations) the majority of the collective dose is likely to be made up of contributions to individuals living outside the country of its origin.

An important aspect of the problem is in the long-term nature of source of release of radionuclides. Untreated tailings piles may be expected to continue to be a source of radiation exposure for billions of years after being laid down.

1.4. Objectives of the Co-ordinated Research Programme

It is evident that there are many aspects to the effective management of uranium mining and milling wastes. This CRP has been directed at studying one particular associated topic area, namely the migration of radium and other contaminants from tailings. It is therefore particularly concerned with the migration and transfer processes caused by ingress of water into tailings piles. Some of the studies in the programme have been concerned with the behaviour of radium after transfer into aquatic systems while others were concerned with the basic processes which occur in tailings piles. Studies of the latter type are essential if the long-term behaviour of radium and other contaminants is to be predicted with confidence and the effects of various treatment schemes are to be understood. The migration behaviour of radium-226, as a precursor to radon-222, will also have an effect on the long-term emission of this pollutant gas.

2. HISTORY AND ORGANIZATION OF THE CO-ORDINATED RESEARCH PROGRAMME

This Co-ordinated Research Programme (CRP) followed one on a similar topic "The Source, Distribution, Movement and Deposition of Radium in Inland Waterways and Aquifers" (1976-80) (IAEA 1984a).

The CRP was started in 1981 and a total of 11 contractants from Argentina, Australia, Brazil, Canada, Czechoslovakia, France, Gabon, India and the USA were finally associated with it (for details see Appendix 2).

There have been three research co-ordination meetings in the course of the CRP. They were held in Ottawa, Canada (May 1982), Rio de Janeiro, Brazil (February 1984) and Cadarache, France (September 1985). Each was attended by local interested scientists in addition to the core of principal investigators from the contracted organizations. The format for the meetings was similar in each case. Reports of progress in the research were made by each of the principal investigators and additional presentations on related work were made by local or other invited experts. Working groups were established to discuss research needs in different subject areas. The results of the discussions of these working groups and their recommendations are recorded in Chapter 3 of this report. A visit to a local mining and milling facility was arranged in association with each each of the meetings and this was also used as a focus for discussion between participants. The facilities visited were as follows: Madawaska mines and tailings, near Bancroft, Canada, Nuclebras mining and milling industrial complex, Pocos de Caldas, Brazil, and the Lodève mining and milling facilities, Herault, France. A report of the discussions on the Pocos de Caldas visit is included in the informal report on the second research co-ordination meeting (IAEA 1984b).

3. DISCUSSION AND RECOMMENDATIONS OF WORKING GROUPS

This section is an edited version of material produced at the third and final Research Co-ordination Meeting in Cadarache. However, it draws on reports produced at the earlier meetings in Rio de Janeiro and Ottawa.

3.1. Source terms

3.1.1. Physico-chemical forms of radionuclides and other contaminants in mining-milling wastes

In the mining and processing of uranium ores, there are a number of possible waste discharges: these wastes, in addition to containing the radionuclides, could also contain (depending upon the ore type, mineralization and process used), other contaminants such as nickel, arsenic, molybdenum, zirconium, selenium, copper, barium, iron, manganese, chloride, fluoride, nitrate, sulphate, ammonium, etc. The wastes include:

- Waste rock from mining;
- Overburden (from open pit mining);
- Mine water;
- Mill process waste residues;
- Neutralized mill effluents;
- Radium-barium sulphate precipitates;
- Ferric hydroxide precipitates;
- Gaseous effluents;
- Organic constituents (solvent extraction reagents, diesel oils, flotation agents, biological constituents, e.g., humic acids, bacteria, algae, fungus).

This chapter deals with the geochemistry of the sources of contaminants as they are discharged, followed by the biogeochemical mechanisms that could occur with exposure to the subsequent weathering process. The aspects of site selection, alternative technologies, and potential options for utilization of these industrial wastes, although important, are not covered in this document. The actual sources have been mentioned by a number of the CRP contributors.

The physico-chemical forms depend on the mineralogy, the process used in the mill, and the treatment of the effluents (e.g., BaCl₂ and/or neutralization processes). (Peterson and Gee, 1985; Raghavayya, 1985; Evans, 1984; Ritcey and Chomyn, 1984; Ritcey and Silver, 1982; Skeaff et al., 1982; Gee, 1984; Franca et al., 1984).

Obtaining an understanding of the physico-chemical mineralogical forms of wastes from uranium mining and milling operators is most important if the prediction of contaminant migration is to be eventually achieved. Very little has been done in this area to date, but at present there are limited investigations in progress. Some reported data are available from IAEA TECDOC-301 (IAEA 1984a) on physico-chemical forms of radium in uranium mining-milling wastes.

The working group recommended that additional data be obtained on the constituents (chemical, biological and mineral) as well as the speciation of radionuclides and other contaminants in waste solution and on solids. More knowledge and data are required on the relationship between speciation and factors such as ore and waste composition, and the treatment process used.

3.1.2. Biogeochemical mechanisms

The geochemical mechanisms controlling release of radionuclides and other contaminants from tailings and waste rock have been investigated by several of the groups contributing to the CRP (Fourcade and Zettwoog, 1985; Ritcey, 1984; Benes et al., 1982-1985; Canet and Mery, 1982; Gee, 1984). The mechanisms include adsorption-desorption (ion exchange), precipitation-dissolution and coprecipitation reactions which are affected by such parameters as pH, Eh, solid surface properties and biological reactions that are often both time and space dependent. A summary of how these mechanisms affect radium release and migration is given in Chapter 5 of IAEA 1984a. Much work remains to be done in order to understand the processes which control radium, and other heavy metal migration from solid wastes subjected to such processes as biological activity, leaching, freeze-thaw cycles and having a variety of site-specific chemical and mineralogical characteristics and compositions. Work on various aspects of the biogeochemistry of wastes performed by CRP members can be found in the following references (Fourcade and Zettwoog, 1985; Ritcey and Silver 1985; Peterson and Gee, 1985; Evans, 1984, 1985; Ritcey and Chomyn, 1984; Skeaff, et al., 1982; Silver 1983; Benes et al., 1982-1985; Canet and Mery, 1982; Fourcade et al., 1983; Fourcade and Canet, 1984; Raghavayya, 1982-1983; Gee, 1984; Peterson et al., 1983; Amaral et al., 1984; Azevedo and Schuettelkope, 1984; Oliveira and Franca, 1984; Oliveira et al., 1984; Franca, et al., 1984b).

Lysimeters in Canada (Ritcey, 1984; Ritcey and Silver, 1982) will provide data for basic mechanistic studies on the biogeochemical interactions occurring within the waste dumps. Further discussion of specific studies on the basic biogeochemical process is given in the section on migration (3.2).

The working group recommended that studies continue which identify mechanisms for controlling radionuclides and heavy metals and other contaminants migration from solid wastes under the range of conditions which exist at typical tailings ponds and waste disposal sites.

In addition, it recommended that expanded efforts should be made to identify the interaction of the biogeochemical processes taking place in the wastes, due to the presence of the bacteria, algae, fungi, and other organics.

3.2. Migration

The ultimate goals of the migration studies are to elucidate the pathways of contaminants from their sources to man, to help in finding suitable methods of pollution abatement and to obtain necessary data for construction of appropriate migration models. In order to make any prediction as to migration into the hydrosphere and lithosphere it is necessary to develop a comprehensive understanding of the basic physico-chemical processes occurring between solids and liquid and to determine the speciation of the liquid and solid phases.

3.2.1. Basic processes

Several of the studies within the CRP have been aimed at evaluating basic processes affecting migration (Evans, 1984; Benes, et al., 1984; Gee, 1984). They include studies of adsorption and desorption of radium on model solids representing components of freshwater sediments

(Benes, Strejc, Luckavec, 1984; Benes, Borover and Strejc, 1984) (ferric hydroxide, quartz, kaolinite, montmorillonite, muscovite, albite and humic substances) and on waste water and riverbed sediments (Benes and Strejc, 1985). Pronounced effects of pH, liquid to solid ratio and solution composition were found. It has been concluded that the adsorption affinity of sediments for radium cannot be easily derived from their composition or other properties. No simple correlation with specific surface area, organic matter, oxidic coatings or other components of sediments was observed. However, an exceptional role of barite (barium sulphate) in the sediments was noted (Benes and Strejc, 1985). K_d coefficients strongly depend on the liquid to solid ratio and other factors and therefore K_d does not represent a parameter suitable for quantitative descriptions of radium interaction with solids in surface waters (Benes, Strejc and Lukavec, 1984; Benes, Borovec, Strejc, 1985a, Benes, Borovec, Strejc, 1985b).

Adsorption on model oxides and clays has also been studied (Lowson and Evans, 1984). It was shown that the pH at which 50% of the original adsorbate is adsorbed (pH_{50}) was a function of ion type and this allowed the value to be interpreted as a measure of the relative affinity of adsorbate for the surface. Below a limiting solution concentration of about 10^{-7} mol. l^{-1} , the pH_{50} value became independent of concentration for a range of cations and an adsorption mechanism involving uptake of the hydrolyzed ion onto active sites accounted for the observations. The approach was concluded to be a convenient treatment of adsorption processes from low concentration solution which merits further evaluation.

The adsorption of As, Mo, Se, and Ra from acidic tailings effluent on alkaline, sandy subsoils has also been studied (Peterson and Gee, 1985).

In addition, mechanisms of coprecipitation, precipitation-dissolution, and peptization have been identified as major processes controlling migration potential for Ra-226 and the basic situation for radium has been reported in some detail in Chapter 5 of IAEA 1984a, and by Benes, Sebesta and Sedlacek, 1984. It would be useful to extend the work to U-238, Th-230 and heavy metals.

The working group recommended that basic physical and chemical processes determining migration should be quantitatively characterized, particularly diffusion, sedimentation, resuspension, adsorption, desorption, precipitation, etc.

3.2.2. Speciation (physico-chemical forms in liquids and solids)

Since the physico-chemical forms (speciation) of radionuclides and other contaminants is the primary factor influencing the migration, these forms should be analysed, and their changes during the migration should be studied.

This was in fact recommended at the 1982 RCM in Ottawa. It was further suggested at that time that suitable analytical methods be developed to evaluate the chemical forms of radionuclides in the liquid and solid phases of the tailings and those sorbed on sediments. The speciation of radium, lead and other metals has now been investigated by several of the groups in the CRP (Evans, 1984; Benes, et al., 1984; Canet, 1983; Peterson, et al., 1983; Teixeira and Franca, 1984).

The working group recommended that additional studies be conducted in this respect with the aim of improving speciation

methods and obtaining more data on the speciation. The next step should be an interlaboratory comparison of speciation methods.

3.2.3. Migration studies in the hydrosphere, lithosphere and atmosphere

The hydrosphere, particularly surface and ground waters, are possibly the most important medium for transportation and concentration of most of the contaminants except radon. Gaseous emission of radon and resuspension of solids have been investigated extensively. This subject is of recognized importance but is outside the scope of this CRP.

Studies that have specifically identified and evaluated migration of Ra-226 include both studies of Ra-226 in surface waters (Benes, et al., 1984, 1985; Franca et al., 1984) and in tailings effluents (Evans, 1984; Ward et al., 1981; Canet and Mery, 1982; Fourcade and Zettwoog, 1984; Raghavayya, 1982, 1983, 1984). Radionuclides, other than Ra-226, that are of interest in migration studies include U-238, Th-230, Ra-228, Po-210 and Pb-210. The migration of U-238, Th-230 and Pb-210 has been studied in some detail (Azevedo et al., 1985; Donaci and Gullucci, 1985; Descamps, 1985; Peterson and Gee, 1985; Evans and Giles, 1985; Ritcey and Silver, 1982, 1985; Ritcey, 1984; Benes et al., 1984, 1985; Gee, 1984; Kalin and Smith, 1984; Azevedo and Schuettelkope, 1984; Oliveira and Franca, 1984; Oliveira et al., 1984; Franca et al., 1984; Canet et al. 1985; Franca et al., 1984), but very few studies have addressed the migration of Ra-228 or Po-210.

Research on heavy metals (e.g., Pb, Cu, Zn, As, Se, Mo, Mn) have been conducted by groups represented in the CRP (Lowson and Evans, 1983; Haw et al., 1983; Ritcey and Silver, 1985; Benes et al., 1984, 1985; Gee, 1984; and Peterson et al., 1983). The migration of heavy metals of concern depends upon the ore mineralogy, the milling process and the waste treatment.

In order that the data on contaminant migration in solutions are to be of value, it is necessary for the biogeochemical characteristics of the receiving soil to be obtained.

The working group recommended that more data be obtained on the radionuclide sorption and other chemical interactions that occur between the various contaminants and the receiving soils. This would include a documentation of the physico-chemical aspects, mineralogy and identification of organic and microbial species present.

K_d data on the sorption on to soils have been produced by many investigators. Although K_d is a convenient factor, its use can often give misleading results when it is applied in conditions other than those for which it was determined.

The working group recommended that care be taken in the application of K_d values for conditions other than those in which the data has been obtained. Ideally, other methods which more realistically represent the physical and chemical processes should be developed. (A similar recommendation is made by the modelling working group.)

3.3. Biological uptake

The subject of biological uptake received only limited attention in the CRP and therefore the following discussion is limited in its scope. The subject received attention in the work of the following:

Amaral et al., 1984, Azevedo et al., 1985, Descamps, 1985, Evans et al., 1984, Raghavayya, 1985.

The most common approach for determining biological uptake is by the use of concentration factor, CF* (the limitations of which will be discussed later in this section).

Results of measurements made on transfer to many plant and animal species show wide ranges of CF values. This variation is due, in part, to the complexity of the biochemical processes involved but may also be a result of the different experimental procedures used.

The biological uptake of radionuclides depends on (amongst other things):

- Physico-chemical form of the radionuclide in the medium (soil, sediment and water);
- Radionuclide bioavailability for uptake by plants and animals;
- Differences between species and within individuals in the same species.

Radionuclide availability for uptake is greatly influenced by the chemical and mineralogical composition of the medium, for example, pH, organic matter content, presence of adsorbed materials and analogous elements. In the case of radium, the presence of clay, iron oxide and organic matter or of competitive ions such as calcium and barium decrease its bioavailability.

However, differences in sampling, measurements and normalization procedures have a great influence on observed CF values. In this respect some of the most important factors are:

- Stage of the growth cycle;
- Sampling representativity;
- Preparation procedures (washing, removal of outer skin, etc.);
- Radionuclide content determination (the total or the bioavailable);
- Accuracy of the analytical procedure;
- Type of units used (based on wet, dry or ash material).

Besides the above mentioned reasons for the variability of CF values there are also problems associated with its use in some situations. The uptake of radionuclides into biological systems is a dynamic process and the CF parameter being only strictly applicable to systems in equilibrium is therefore not universally applicable. For many systems the biological uptake mechanism is not well known and for example the variation of CF with the concentration of the radionuclide in the medium is often not understood. For these reasons, care has to be taken in applying the concentration factor approach.

Future work

The extent to which transfer in a particular system should be studied depends upon the objectives of the research or assessment

* CF = $\frac{\text{Radionuclide concentration in biota}}{\text{Radionuclide concentration in the supporting medium}}$

programme. If the studies are being conducted for the purpose of ensuring the protection of man, it should be determined which pathways are most significant in relation to the exposure of man by means of a preliminary modelling exercise. In this way it may be possible to avoid costly and sometimes unnecessary studies of transfer into systems which are comparatively unimportant from a radiological point of view.

The working group considered that the emphasis in future work in this subject area should be as follows:

- On developing analytical methods for the determination of the bioavailable radium in supporting materials;
- On studying, in greater detail, transfer mechanisms in the pathways to man which have been established as the most important concerning the radiation exposure to man;
- In attempting to standardize the sampling and preparation procedures as well as the methods for normalizing results.

3.4. Modelling Aspects

In this working group modelling was considered mainly in relation to the need to assess the impact on man of radium and other contaminants from mining and milling operations. Such modelling is often termed "assessment modelling" (as distinct from research modelling). In the CRP studies of both types of modelling were reported (Peterson et al., 1985; Baptista et al. 1985; Benes et al., 1985; John, 1985).

The discussion focussed on the information needs for effectively modelling migration and transfer to man. Several of the points raised by the other groups were also identified in relation to data and research needs for modelling.

Numerous potential pathways to man have to be considered, and for the sake of simplicity they are grouped under three headings. It is recognized, however, that many pathways are interconnected and may occur under headings other than those under which they are considered here. The headings are:

- | | | |
|-------------------------|---|---|
| Hydrogeochemical models | - | includes source terms, with ground and surface water pathways |
| Air dispersion models | - | includes both radon and dust migration |
| - | | |
| Sedimentation models | - | mechanisms for removal of radionuclides from the system |

Hydrogeochemical modelling

The working group considered that more research should be carried out on the following topics:

1. On acquiring more data on the solubility of minerals and solid phases;
2. On the adsorption of various contaminants onto pure solids and non-heterogeneous mixtures of solids;

3. On the validity of scaling dispersion coefficients to conditions other than those in which they were obtained;
4. On the use of probabilistic modelling to predict water flow.

The distribution coefficient (K_d) approach to modelling adsorption is deficient in many respects and it is desirable that other approaches be developed. Other approaches which warrant further investigation include surface complexation models that intrinsically account for changing solution parameters such as pH, ion strengths, solution composition and changing electrostatic potentials at the solid-liquid interface.

Air dispersion modelling

This subject has been well researched and a number of reliable models exist which can be used directly or fairly easily adapted. Further development of these models need not receive a high priority except for very specific needs.

Sedimentation modelling

There is a lack of understanding of the fundamental mechanisms associated with the movement of radionuclide through the sediment system. Radionuclides can enter the sediments in many ways, for example as precipitates with either organic or inorganic chemicals; as part of the body of dead animal or plant tissues, or by absorption, adsorption or ion exchange. Once in the sediment the radionuclides will eventually become buried by other natural deposits. Until they are buried below the active layer radionuclides will remain available for remobilization. Again there are several potential mechanisms such as chemical reaction and leaching; bioperturbation and erosion.

To date most of these mechanisms are not well understood. The extremely variable nature of sediments and surface water systems, coupled with the complex biological environment will make the development of such an understanding difficult. It is to be hoped that current research will provide simple component models which will give a reasonable approximation to the real system.

In addition, the distributions of the input parameters are poorly known and difficult to measure. Parameters, such as sedimentation rate, are subject to a variety of environmental influences and the collection of enough data to define the uncertainty associated with the parameters is expensive and time-consuming.

The working group recommended that to improve the possibilities of sedimentation modelling research into sedimentation processes and parameters be given a high priority.

For all types of model it is desirable for research on validation to be carried out, that is the testing of model predictions against real and appropriate experimental data. Other techniques for increasing confidence in model predictions should also be pursued since complete validation is rarely possible; these include model comparison and statistical techniques.

3.5. Conclusions

Until the physico-chemical processes governing the interaction and migration of heavy metals and other pollutants present in the wastes from uranium mining and milling are properly understood, there will continue to be considerable uncertainty associated with predictions of their associated radiological and other possible health impacts. The importance of this research is recognized, especially in the Member States where uranium is being mined and processed and considerable efforts are continuing to be applied to the subject.

The discussions and exchanges of information which this CRP has made possible have contributed towards an increase in the general understanding of the subject. The CRP has served to focus attention on the most important of the unresolved issues and has stimulated greater research efforts in these areas in individual Member States. However, as may be seen from the foregoing sections, there remains much to be done if a sufficient understanding is to be obtained.

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APPENDIX 1

GUIDANCE ON ANALYSIS AND MEASUREMENT PROCEDURES (assembled at the 1982 Ottawa meeting of the CRP)

1. INTRODUCTION

The environmental impact of the mining and milling of uranium has not been clearly evaluated. Models are needed which can predict the long-term consequences of large volumes of radioactive materials stored near the earth's surface. These tailings materials will be in contact with varying quantities of water, which over time may leach contaminants to surface and groundwater and will also be subjected to biological forces (plant and animal intrusion). As a consequence of these processes radioactive material may ultimately be transferred to man via his dietary intake.

In order to properly model this system and to make adequate predictions for long-term containment of these wastes, it is clear that the models need to be built upon a clear description of the chemical, biological and physical processes that occur in the tailings and upon accurate data sets obtained from reliable analysis.

The purpose of this Appendix is to describe appropriate analytical procedures for radioactive contaminant analysis of tailings. This is an essential first step before any long-term impact assessment can be attempted.

2. ANALYTICAL

2.1. Sampling Procedures

The first essential to a successful monitoring or field investigation program is the use of correct sampling procedures which provide a representative sample for analysis and the design of a sampling protocol to meet the needs of the particular site.

2.1.1. Water

a. Sampling Procedures

Sample types - Grab
- Composite
- Continuous

Sample bottles - polyethylene or polypropylene (glass is preferable if ^{210}Po analysis is to be carried out)

b. Sample Treatment

Only simple water chemical tests such as conductivity, Eh and pH, are normally carried out immediately on sampling. For more sophisticated tests, samples are transported back to a laboratory. In the ensuring period between sampling and analysis, changes can occur in the sample by adsorption on the vessel and redistribution between phases which can affect the final result. To minimize the effects the solid phase should be separated in the field, the samples acidified and stored at low temperatures.

Separation

Filtration - normally a membrane filter such as Millipore is used. Nucleopore provides a more specific sample size fractionation but is slower. Their main application is in speciation studies.

Centrifugation - used to abstract the solids from a large volume of water to provide a useful sized sample.

Ultrafiltration (UF) - used to obtain sample of colloid-size particles. Care must be taken to check that significant adsorption is not occurring on the large surface area membrane.

Dialysis (in-situ) - A dialysis bag immersed in the water for two days to allow equilibrium will provide a representative sample of the ionic species present.

Preservation

Add acid to sample to bring up to 10^{-2} mol L⁻¹; HCl or HNO₃ may be used depending on the analysis to be carried out.

Should not be used for speciation studies.

Freezing is sometimes used but may change the form of colloids if speciation studies are intended.

c. Storage

Samples are stored at low temperature (4°C).

2.1.2. Solids

a. Sample

The sample must be representative and should be at least 1 kg.

b. Sampling Protocol

Design of the sampling plan is site-specific and requires a previous investigation with depth profiles of soils and sediments.

c. Sample Treatment Prior to Analysis

Requires homogenizing.

The sample will usually be dried (commonly 105°C) but the choice depends on the analysis to be carried out. For ²¹⁰Po low temperature is used. If speciation studies are to be carried out, the effect the drying should be investigated.

d. Sample Storage

Wet samples should be stored at 4°C.

2.1.3. Interstitial Water

Interstitial water may be separated from sediments, soil and tailings by various techniques:

1. Centrifugation
2. Displacement by an inert immiscible organic liquid, e.g. fluorinated hydrocarbon
3. Dissolved by a miscible organic liquid, e.g. alcohol, acetone
3. Dialysis in situ (sediments)

2.2. Methods of Measurement

Introduction

Only the main methods of measurement in current use are described here. Reference should be made to more comprehensive analysis manuals, e.g. Volchok and de Planque, Smithson, et al., and Dean & Chui (1985).

2.2.1. ^{226}Ra

a. Emanometry

Radon emanation is the most widely used method and is used in several techniques to analyse for ^{226}Ra via its daughters. Lucas-type cells or similar are frequently used for low-level water samples. However, attention must be given to particulates. Instead of de-emanation, the radon can be extracted into an organic solvent and measured via liquid scintillation. In the case of the higher activity solutions, e.g. wastewater, these can be extracted in the scintillation vessel and counted without phase separation.

b. α -Spectroscopy

α -spectroscopy using the 4.78 MeV peak is sometimes used. The radium is usually separated in the form of Ba-Ra sulphate. Alternatively, a thin deposit is applied to MnO_2 . An efficient plating technique capable of operating in the presence of high calcium concentrations would greatly widen the scope of the method.

c. γ -Spectrometry

γ -spectrometry provides a direct analysis using the 186 keV ^{226}Ra peak. However, when using this line, possible presence of ^{235}U or ^{228}Ra must be taken into account. Alternatives to the use of the 186 keV line are the ^{214}Pb or ^{214}Bi peaks, e.g. 295, 352, 609, 1120 or 1760 keV, but in these cases the possibility of disequilibrium between $^{226}\text{Ra}/^{222}\text{Rn}$ must be checked.

With NaI(Tl) detectors, special attention must be given to $^{226}\text{Ra}/^{228}\text{Ra}$ daughter product peaks overlapping.

d. α -Gross Counting

This is not a selective method in the way that the emanometric method is. Interferences may arise due to ^{223}Ra , ^{224}Ra and ^{228}Ra or daughter products. But, if

correctly used it can be considered as an alternative to the emanometric method.

e. Other Methods

The use of β - γ or α - γ coincidence methods for the ^{226}Ra determination is increasing.

2.2.2. Other Radionuclides

- ^{228}Ra : Measurements through ^{228}Ac - β - γ coincidence, γ -spectrometric or β -counting
- ^{210}Pb : Low-energy γ -spectrometry, β -counting (^{210}Bi), γ -counting (^{210}Po)
- ^{210}Po : α -counting or α -spectrometry
- $\text{Th}_{\text{nat.}}$: Activation analysis, spectrophotometry, α -spectrometry
- ^{230}Th : α -spectrometry
- ^{228}Th : α -spectrometry or γ -spectrometry
- $\text{U}_{\text{nat.}}$: Fluorimetric, activation analysis, γ -spectrometry, α -spectrometry, spectrophotometry
- ^{234}U : α -spectrometry
- ^{235}U : α -spectrometry or γ -spectrometry

2.3. Speciation Procedure

For the purpose of pathway analyses of radionuclides, total radionuclide analyses alone are insufficient and therefore the water sample must be fractionated into its component parts to identify the species present. Similarly, with solids, a sequential leaching procedure is used to separate the mineral components.

Water Phase

Dissolved species and particulates are characterized following separation by filtration, centrifugation, ultrafiltration or dialysis.

1. Dissolved

a. In true solution

Metal complex ions (chloride, carbonate, sulphate), simple metal ions (Ra^{2+})
Molecules, uncharged (Low molecular weight organic complexes)

Methods

- electrophoresis
- diffusion
- ion exchange
- solvent extraction
- co-precipitation

b. Colloids - lyophilic, lyophobic

Methods

- centrifugation
- oxidation (to destroy organics)
- ultrafiltration
- gel filtration
- electrophoresis (for characterizing)

2. Particulates

Co-precipitates (SO_4 , CO_3)

Adsorbed

- clays
- oxides (Mn, Fe)
- carbonates, etc.

Method

- sequential extraction

3. Sediments/Soils/Tails

Method

- sequential extraction

APPENDIX 2

SUMMARIES OF RESEARCH CARRIED OUT BY INDIVIDUAL CONTRACTORS

Research Contract No. 2890/CF, Chief Investigator: J.W. Evans,
Environmental Chemistry Section, Lucas Heights Research Establishment
Australian Atomic Energy Commission, Lucas Heights, AAEC Private Mail
Sutherland, NSW 2232, Australia

Title: Studies related to the environmental migration of radium

The AAEC programme covered by the IAEA contracts consists of biological uptake studies of radium by macrophytes, mussels and turtles in a tropical environment, investigation of radium concentration in high organic soils of the Northern Territory of Australia and more basic chemical studies of solution and adsorption processes active in geochemical transport.

Although not part of this CRP, because of their interest to the group an account is also given of research on tailings chemistry at the AAEC and on radium in groundwaters at CSIRO (Commonwealth Scientific and Industrial Research Organization).

The titles of the main study areas are listed below:

1. BIOLOGICAL UPTAKE STUDIES
 - 1.1. Radium-226 Uptake by Waterlily, Nymphaea.
 - 1.2. Role of Macrophytes in Radium Cycling on the Magela Floodplain.
 - 1.3. Uptake and Loss of Radium-226 by Freshwater Mussels.
 - 1.4. Radium-226 Kinetics in Freshwater Turtles.
2. GEOCHEMICAL TRANSPORT
 - 2.1. Chemical Processes
 - 2.1.1. Solution chemistry
 - 2.1.2. Adsorption studies
 - Cation Adsorption
 - Role of Colloids
 - 2.1.3. Concentration of radium in swamp soils
 - 2.2. Chemistry of Uranium Mine Tailings
 - 2.2.1. Geochemistry of radionuclides
 - 2.2.2. Iron chemistry in tailings
 - 2.2.3. Rum Jungle Tailings
 - 2.2.4. Dewatering of tailings
 - 2.2.5. Long-term leaching of tailings
 - 2.3. Radium Chemistry
 - 3.3.1. Thermodynamics
 - 3.3.2. Solution chemistry
 - 3.3.3. Analytical
3. RADIUM IN GROUNDWATER (CSIRO)

Research Contract No. 2891/CF, Principal Investigator: G.B. Baptista, Pontificia Universidade Catolica do Rio de Janeiro, Departamento de Fisica, Rua Marquês da Sao Vicente 225, Gávea, Rio de Janeiro, RJ 22453 Brazil

Title: Ra-226 collective dosimetry for surface waters in the uranium mining region of Pocos de Caldas

Work had been carried out on this topic prior to the period of the CRP. However, during the course of the CRP, studies had to be suspended for reasons outside the control of the research team. The research moved to related subjects of a more general and theoretical nature. For example, information on the behaviour of radium in the environment was used to derive environmental reference standards for radium-226 releases based on ICRP dose limits. Other work has included an investigation of the particle size distribution of yellowcake in the aerosols produced in routine operations and subsequently estimation of radiation doses to workers as a result of deposition of uranium in the respiratory tract.

Research Control No. 2892/CF, Chief Investigator: G.M. Ritcey, CANMET, Department of Energy, Mines and Resources, 555 Booth Street, Ottawa, Ontario, Canada K1A 0G1

Title: Simulated Tailings Weathering Tests in Lysimeters

During the past 6 years there has been continuing research progress with CANMET on the study of the reactions occurring within uranium tailings. These investigations have been carried out in lysimeters, or controlled test containers, in which accelerated simulated weathering has taken place. The initial work was on Elliot Lake tailings and the seepage effluents were measured as to the changing chemical compositions over time.

The results of the initial test, which were carried out at a 9-fold simulated weathering cycle (based on rainfall), indicated the following:

- (a) Migration of radium was downwards;
- (b) As the sulphate was depleted, the solubility of radium increased;
- (c) In the presence of oxidizing bacteria, Thiobacillus ferroxidous, sulphides were converted to H_2SO_4 ;
- (d) Tailings become acid (pH 2.5-3.5) after 3-simulated years in the presence of bacteria;
- (e) The presence of a bactericide inhibits the rate of pyrite oxidation;
- (f) The rate of migration of amine and kerosene (present if a solvent extraction purification process is used) showed 2 years and 11 years respectively for their dissipation.

In other tests at CANMET on the use of $BaCl_2$ as a precipitant for radium, although proven effective for significantly lowering the radium concentration in seepage, the barium-radium sulphate could eventually break down if reducing bacteria are present - thus releasing both radium and barium into the environment.

The lysimeters studies have continued and have been expanded to include the chemical, physical, biogeochemical, and geochemical interactions occurring with time for different types of tailings.

As part of the simulated weathering tests performed in lysimeters, concurrent data were obtained by water leaching the tailings, with the effect of limestone and/or peat mass sorption of radium also being investigated. In all the tests on the various tailings, the data indicated that the inclusion of both limestone and peat moss below the tailings in the columns resulted in a substantial decrease in the concentration of radium-226 in the effluents from these columns. Radium may also be removed by passing the effluents through limestone and wetland vegetation. These data suggest that such readily available materials may be practical and economical for the removal of radium from uranium tailing effluents.

The effects of various cover materials on the inhibition of pyrite oxidation by the exclusion of oxygen and the promotion of grass growth have been investigated. Tests on tailings submerged under water were also conducted to determine whether oxidation of sulphides would occur. The results of these investigations suggest that sulphuric acid generation resulting from bacteria pyrite oxidation in uranium tailings can be completely arrested by either submerging the tailings under water, or by the deposition of an organic layer on the surface of the tailings.

Ongoing Research

Complete elimination of sulphuric acid generation by either a layer of water or a sufficiently thick layer of organic material suggests that these procedures may be useful as effective and economical methods of controlling sulphide mineral oxidation and therefore acid seepage from pyritic tailings. Subsequent research is currently in progress to determine the merits of depositing a layer of vegetation obtained from typical Canadian bogland to eliminate acid seepage from pyrite-containing tailings.

Lysimeters to simulate underwater storage of tailings are presently in operation in which deep lake disposal will be simulated in columns 30 feet high and 30 inches in diameter. The quality of the water at various depths will be monitored.

Finally the various aspects of the interactions within the tailings (chemistry, biogeochemistry and physical aspects) have been measured under controlled conditions in a large 11 ton lysimeter. Data on radiochemistry (surface chemistry), clay chemistry and physical properties, magnetic properties and mineralogical changes are presently being accumulated and correlations with migration rates will be made.

Research Contract No. 2893/RB, Chief Investigator: P.Benes,
Department of Nuclear Chemistry, Technical University of Prague,
Prague, Czechoslovakia

Title: Environmental Migration of Radium and Other Contaminants
Present in Liquid and Solid Wastes from the Mining and
Milling of Uranium

The research represented continuation of earlier work carried out in the frame of CRP "Radium 1" (1976-1980) and had three principal objectives: to elucidate the interaction of radium with freshwater sediments, to analyse the radium bond in uranium mill tailings and to study the migration of ^{210}Pb and stable lead in surface waters affected by uranium mining.

A series of laboratory experiments was carried out on the adsorption and desorption of radium on riverbed and waste water sediments and on model solids representing components of the sediments: ferric hydroxide and quartz, kaolinite and montmorillonite, muscovite and feldspar (albite). The effects of pH, liquid to solid ratio, time, ionic strength (Na^+) and presence of calcium and sulphate ions were studied under conditions simulating as closely as possible the conditions prevailing in surface (continental) waters. Conclusions were drawn on the mechanism of radium interaction with the studied solids, on the importance of the solids for radium migration in surface waters and on the effect of ambient factors on the interaction of radium with freshwater solids. It has been found that the adsorption affinity of sediments for radium cannot be easily derived from their composition or other properties. No simple correlation with specific surface area, organic matter, oxidic coatings or other components of the studied sediments was observed. However, an exceptional role of barium sulphate in the sediments was noted.

The radium bond was analysed in four different samples of dry uranium mill tailings using a method of selective dissolution developed earlier. It was found that three of the analysed samples contained radium predominantly in "acid soluble" form, representing the radium bound in oxidic or carbonate components of the tailings or strongly adsorbed on minerals. Only one sample (of South African origin) contained radium mainly in "crystalline detritus", i.e. radium incorporated in the crystalline lattice of silicate minerals. The results have proved that the method of selective dissolution can provide useful information on radium bonding in waste solids.

Migration of stable lead and ^{210}Pb was studied in two surface water systems representing two main sources of local contamination of the hydrosphere from uranium mining: mine drainage waters and spoils (waste rock) piles. In one system mine drainage waters treated by precipitation of barium sulphate contained considerable activity of ^{210}Pb in particulate form. Spoils piles were the main source of ^{210}Pb in the second system with dissolved ^{210}Pb being the predominant migration form. The highest concentration of dissolved ^{210}Pb here was found in a creek under low flow conditions, pile seepage water contained generally low concentrations of ^{210}Pb . In both systems the concentration of ^{210}Pb rapidly decreased with distance from the source indicating that waterborne ^{210}Pb did not represent an environmental problem. Uranium mining did not add significant amounts of stable lead into the surface water systems. It has been found that bottom sediments are a sensitive indicator of the pollution of surface waters with ^{210}Pb .

The study was supplemented by the analysis of the bond of ^{210}Pb in bottom sediments of the first system. The method of selective dissolution was suitably modified for this purpose. It was found that a significant part of ^{210}Pb was present as "crystalline detritus" and "acid soluble" forms in the sediments. The nature of ^{210}Pb distinguished as being in the "barium sulphate" form requires further study.

Research Contract No. 2894/RB, Chief Investigator: P. Zettwoog,
Service de Protection des Installations Nucléaires, Department de Protection
(IPSN), CEN-FAR, B.P. No. 6, F-92260, Fontenay-aux-Roses, France

Title: Storage of the wastes from the extraction and treatment of uranium minerals in France

The research programme is essentially aimed at obtaining the necessary environmental data to allow the radiological impact of effluent discharges arising from the uranium mining and milling activities in various areas of France, principally, Vendée, Limousin and Lodève to be assessed. A model has been developed which allows the individual and collective radiation doses to be assessed due to the transfer of radium-226 via soils and plants as a result of transport by rivers or sub-surface water of radionuclides from the mining and milling sites.

An important part of the studies has concerned the evaluation of the fractions of the total radium present at various stages of the mining and milling process which are capable of being solubilized by water.

Research Contract No. 2895/RB, Chief Investigator: M. Raghavayya,
Health Physics Unit, Bhabha Atomic Research Centre, Indian Rare-Earths Ltd
Udyogamandal 683501, Kerala, India

Title: Study of the mechanisms of Ra-226 and radon daughters release in the environment of uranium mining and milling

The physico-chemical properties of the mill tailings at Jaduguda were studied (particle size, radium content etc.). Fractionation studies indicated that about 0.5% of the input radium becomes effectively solubilized at the end of the process. Leachability of radium in the tailings was determined using rain water, distilled water and also radium depleted tailings effluent. Only about 0.5% of radium was solubilized with these media. About 13% of radium in the tailings particles could be leached with a 400 ppm chloride solution. 66% of radium could be leached with 0.25 M EDTA at pH 9. Other organic solvents were less efficient at leaching radium.

In a different part of the country, the $\text{CaCO}_3 + \text{CaSO}_4$ sludge from a phosphate based fertilizer plant was also similarly studied since it contains about 1.1 Bq of radium per gram. Rain water in this case, since its pH is low (4.5), could leach out more radium. With a continuous column two states of radium in the sludge could be inferred - loosely bound and chemically exchangeable.

Uptake of radium by vegetable plants was studied. Two types of plants - Hibiscus Esculentus (Lady's Finger or Okra) and Lycopersicum Esculentum (Tomato) were chosen. Plants were grown in soil containing varying amounts of radium and calcium and irrigated with relatively

radium-free water. Radium uptake was observed to be a function of the ratio of available radium to available calcium in soil (inverse proportionality).

Radon emanation from the tailings pond was studied. The rate of emanation ranged from 0.1 to 30.9 Bq.m⁻².sec⁻¹ with a median of 1.5 Bq.m⁻².sec⁻¹. Study of the spatial distribution of radon around the tailings dump indicated that the atmospheric radon concentrations decrease to background levels within a few hundred metres of the periphery of the dump. Emanation of radon from the fertilizer sludge was lower, with a mean of 0.28 Bq.m².sec⁻¹.

Research Contract No. 3018/CF, Principal Investigator: A. Grauby,
Service d'Etudes et de Recherches sur l'Environnement (SERE)
IPSN/DERS, CEN Cadarache, F-13115 St. Paul-lez-Durance, France

Title: Radioecological studies in the aquatic environment of the mining complex of Lodève, France

The objectives of the studies have been twofold:-

To determine the concentrations in water, sediments plants and fish of radium-226, other γ -emitters in the uranium-238 and thorium-232 chains, beryllium-7, potassium-40 and also fission products (Caesium-137, etc.)

To calculate the concentration factors between different compartments of the aquatic environment and to compare them with those obtained elsewhere in France and in other countries.

Eventually, this collection of data, obtained both from the environment and from the laboratory, is expected to make a significant contribution to the data base necessary to allow the assessment of radiation doses to the population.

Research Contract No. 3123/RB, Chief Investigator: B.A. Donati
CNEA, Complejo Minero Fabril, Sierra Pintada, C.P. 5600, San Rafael-Mendoza
Argentina

Title: Research work on radium and uranium migration in Sierra Pintada's Uranium District

The Contract 3123/RB signed between the IAEA and the Atomic Energy Commission of Argentina (CNEA) has the purpose of studying the migration of Ra-226 and Natural Uranium from the San Rafael mining district to the environment. The study area includes the Sierra Pintada uranium district and its zone of influence, both located in the San Rafael Department, Province of Mendoza.

The present study is intended to set precedents for application to other similar mining districts, for mines which are in the stage of production or are already exhausted. The geology of the region, the mining operations, the chronology of the mine production as well as the location of the probable sources of contaminants and the probable paths for the migration of radionuclides have been investigated. Vegetation and fauna species were recorded and samples from vegetation and the main crops in the area were selected in order to obtain background values of Ra-226 and natural uranium. With the same purpose in mind some statistical

analyses were performed in sediments and groundwater samples. Finally, in order to get information on the physico-chemistry of the migration process, some preliminary quasi-quantitative studies were carried out on the most common elements present in surface groundwaters.

From the different evaluations it can be concluded that the level of migration of Ra-226 and natural uranium is low, and from the first measures, no incorporation was observed in vegetables or in manufactured products.

The final recommendations are that it is advisable to continue with this type of study taking samples from vegetation, animals, soils, air, surface and groundwaters with the purpose of controlling possible areas of contamination and areas of specific radionuclide accumulation.

Research Contract No. 3143/CF, Chief Investigator: G.W. Gee,
Geosciences Research and Engineering Department, Pacific Northwest
Laboratory, Battelle Memorial Institute, Box 999, Richland WA 99352,
USA

Title: Studies on environmental aspects of uranium recovery operations

A wide range of environmental aspects of uranium recovery operations has been studied in the United States during the past five years. At the Pacific Northwest Laboratory in Richland, Washington, funding from the U.S. Nuclear Regulatory Commission has provided for extensive evaluation of such diverse subjects as long-term and interim stabilization of tailings piles, tailings dewatering and neutralization, tailings disposal methods, tailings leachate chemistry, radon control, and methods for control of contaminant migration from in situ mining operations. Since 1984, little money has been available to continue much of this work.

Recently, however, progress has been made in several research areas. Three areas of note are: (1) the interactions of ponded acidic uranium mill tailings solutions with indigenous sediments and geologic liner materials, (2) the effects of neutralization on the acidic uranium mill tailings solutions, and (3) the study of methods to minimize groundwater contamination from in situ leach uranium mining.

The objectives of these programmes are summarized below.

Mill Tailings Leaching (Geochemical Aspects)

The principal issues addressed by the experimental and geochemical modelling working under ponded acidic uranium mill tailings are:

- What are the short- and long-term mineralogical and hydrological changes that occur when acidic uranium mill tailings contact sediments?
- Which contaminants will migrate from uranium mill tailings impoundments?
- What are the sediment/solution interactions that attenuate movement of contaminants?
- If the contaminants do move, what will their concentrations be in the migrating solutions and in the groundwater systems?

Tailings Neutralization Studies

In the area of neutralization, laboratory experiments were performed to compare the effectiveness of limestone (CaCO_3) and hydrated lime [$\text{Ca}(\text{OH})_2$] for improving waste water quality through the neutralization of acidic uranium mill tailings liquor. The experiments were designed also to assess the effects of three proposed mechanisms - carbonate complexation, elevated pH, and colloidal particle adsorption - on the solubility of toxic contaminants found in a typical uranium mill waste solution. Of special interest were the effects each of these possible mechanisms had on the solution concentrations of trace metals such as Cd, Co, Mo, Zn, and U after neutralization.

In Situ Uranium Recovery

Our work in in situ leach uranium mining has focused on the early detection of a loss of control of the leaching solution during mining (an excursion) and on aquifer restoration at the termination of mining. Samples of groundwater, leaching solution, and aquifer sediments were collected at mine sites in Texas and Wyoming. These materials were used in laboratory experiments to identify practical indicators of an excursion and to evaluate restoration methods currently in use or considered for future use.

Research Contract No. 3441/RB, Chief Investigator: E.C. Da Amaral
Department of Environmental Radiological Protection
Instituto de Radioprotecao e Dosimetria, Av. Das Americas, km 11.5,
Rio de Janeiro 22700 RJ, Brazil

Title: Environmental studies in the surroundings of the Brazilian uranium mining and milling sites

The first Brazilian uranium mine and mill are located on the Pocos de Caldas plateau, in the Central State of Minas Gerais. The pre-operational environmental survey was carried out over a period of two years by the Brazilian Nuclear Energy Commission (CNEN), through the Instituto de Radioprotecao e Dosimetria (IRD). The selection of the materials to be monitored, the sampling points and the radionuclides to be analysed were based on critical parameters taking into account the meteorological and hydrological characteristics of the site as well as land use. The results obtained are sufficient to characterize the environmental background of the area, and will allow an assessment of the environmental impact due to the operation of the facility and an evaluation of the adequacy of the effluent emission control measures. In relation to their contribution to the population dose, the results indicated that ^{226}Ra and ^{210}Pb will be the important radionuclides.

Research Contract No. 3432/RB, Principal Investigator: M.M. Avome Nze,
Université Omar Bongo, Faculté des Sciences, Laboratoire de Spectrométrie
Nucléaire, BP 911, Libreville, Gabon

Title: Study of daughter products of uranium and their transfer into the environment in the area of the Oklo uranium mine, radiological impact on the population

This project is still in its early stages. Emphasis to date has been on equipping the laboratory with appropriate spectrometers, etc., to allow the assessment of environmental samples.

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