BIOPROTA

Key Issues in Biosphere Aspects of Assessment of the Long-term Impact of Contaminant Releases Associated with Radioactive Waste Management

International Workshop on the Environmental Behaviour of Radium

WORKSHOP REPORT

K Smith, G Smith & Y Thiry (editors)

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PREFACE

BIOPROTA is an international collaboration forum which seeks to address key uncertainties in the assessment of radiation doses in the long term arising from release of radionuclides as a result of radioactive waste management practices. It is understood that there are radio-ecological and other data and information issues that are common to specific assessments required in many countries. The mutual support within a commonly focused project is intended to make more efficient use of skills and resources, and to provide a transparent and traceable basis for the choices of parameter values, as well as for the wider interpretation of information used in assessments. A list of sponsors of BIOPROTA and other information is available at www.bioprota.com.

The general objectives of BIOPROTA are to make available the best sources of information to justify modelling assumptions made within radiological assessments of radioactive waste management. Particular emphasis is to be placed on key data required for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge to the environment or release from solid waste disposal facilities. The programme of activities is driven by assessment needs identified from previous and on-going assessment projects. Where common needs are identified within different assessment projects in different countries, a common effort can be applied to finding solutions.

This report describes presentations and discussions held during an international workshop on the environmental behaviour of radium-226 (Ra-226) on 4-5 May 2010, hosted by ANDRA at their offices in Châtenay-Malabry, France. The workshop was financially supported by ANDRA, NUMO (Japan), NRPA (Norway), NDA/RWMD (UK) and SKB (Sweden). Technical support was provided by a wide range of organisations via presentations and discussions as described in the report.

The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved. All material is made available entirely at the user's risk.

Version History

Version 1.0: Draft report prepared by RPS P&D, ANDRA and GMS Abingdon following the workshop and distributed 02 June 2010.

Version 2.0: Final report prepared by RPS P&D, ANDRA and GMS Abingdon in light of comments received from participants on Version 1.0 and distributed 24 June 2010.

EXECUTIVE SUMMARY

This report describes presentations and discussions held during an international workshop on the environmental behaviour of radium-226 (Ra-226) on 4-5 May 2010 The workshop participants wish to express their gratitude to ANDRA for hosting the meeting at their offices in Châtenay-Malabry, France. The workshop was supported financially by ANDRA, NUMO (Japan), NRPA (Norway), NDA/RWMD (UK) and SKB (Sweden) and this support is gratefully acknowledged.

The workshop involved a number of presentations relating to the behaviour of radium in soils, uptake into plants and approaches to modelling radium transport in biosphere assessments. From these presentations a number of gaps and uncertainties were identified:

- Data relating to the accumulation of radium in soil, especially in clay soils and organic soils, are limited, including thermodynamic data for interactions of radium with organic matter, which limits the ability to calculate uptake into the food chain, but also limits the capacity to estimate external and inhalation doses.
- Data relating to the accumulation of radium in sediments are limited, with similar implications to the above.
- Data relative to the concentration of radium (and daughters) in natural environments.
- Environmental change may affect radium mobility. For example, changes in redox conditions or interactions with barium can have implications for the bio-availability of radium such that the vertical pattern of soil properties could be altered, thus affecting the results obtained from models with multiple soil layers. For near surface disposal facilities, the use of concrete could affect interactions between calcium and radium and such implications could warrant further consideration.
- Transfer to crops is highly variable and is dependent upon both soil and plant properties. It was suggested that soil accumulation and uptake to crops are processes that are overly-simplified. The range of soil to plant transfer factors is very large and there is a need to understand the considerations driving these differences to ensure appropriate parameter values can be selected for specific sites.

Data gaps and uncertainties associated with radium have implications for the distribution of the daughter products Pb-210 and Po-210, which have the potential to dominate radiological impacts due to their high radiotoxicity.

In order to address these gaps and uncertainties, various work programme suggestions were made including:

- Further investigation of factors driving observed differences in soil to plant uptake;
- Appropriate application of analogue elements to address gaps and uncertainties in radium fate in soil-plant systems;
- Review of correlations between environmental concentrations of radium and concentrations in human organs and tissues, including teeth;

- Identification of the key organic matter functional groups controlling radium behaviour in soils to support effective Kd determinations;
- Analysis of clay soil profiles to identify soil gradients that may affect radium behaviour in soils above near-surface disposal facilities;
- Investigation of the effect zones on dose calculations of including multiple soil layers to represent root distributions in soils;
- Review of Biosphere Dose Conversion Factors for different sites to identify features and processes driving differences in results at a site-specific level;
- Investigation of minimum containment requirements for near-surface disposal facilities that meet human and environmental safety criteria; and,
- Investigation of the effect of climatic and environmental changes on the containment of radium in clays and mobility in soils for typical ecosystems.

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CONTENTS

PREFACE **2**

EXECUTIVE SUMMARY **3**

1. INTRODUCTION **7**

1.1 Objectives and scope of the workshop 7

1.2 Participation 7

1.3 Report structure 7

2. PRESENTATIONS **8**

2.1 Overview of data interests based on example assessments for waste management (Graham Smith) 8

2.2 Overview of the effect of phosphogypsum amendments in agricultural soils (Rafael Garcia-Tenorio) 9

2.3 Review of the radiological characteristics of the environment around a former uranium mine site in France: The case of the Saint Pierre site in the Cantal district of France (Marie-Odile Gallerand) 11

2.4 Uranium and its decay products in the human food chain (Marion Jeambrun) 12

2.5 Radium mobility and cycling in re-vegetated mining debris (Yves Thiry) 13

2.6 Radium sorption on soils (Graham Smith on behalf of Abdesselam Abdelouas) 15

2.7 The behaviour of radium and its progeny in mammals (Mike Thorne) 15

2.8 Estimation of soil to plant transfer factor of radium-226 (Keiko Tagami) 19

2.9 Predicting radium availability and plant uptake: Proposal for new best estimates of Kd and TF for radium (Hildegarde Vandenhove) 22

2.10 Environmental behaviour of radium-226 in aquatic and terrestrial ecosystems (Rachid El Mrabet) 27

2.11 Data requirements to determine effective distribution coefficients for radium in soils (Lara Duro) 30

2.12 Ra-226 in a Swedish safety assessment (Sara Norden) 33

2.13 Treatment of radium in the Andra biosphere impact assessment (Achim Albrecht) 36

2.14 Technical Solutions for the implementation of the FAVL disposal concept (Fanny Gerard) 38

3. KEY DATA GAPS AND UNCERTAINTIES **40**

4. DISCUSSION AND IDEAS FOR RESOLUTION OF KEY DATA GAPS AND UNCERTAINTIES **41**

4.1 Soil to plant transfer 41

4.2 Analogue applications 41

4.3 Review of correlations between radium in the environment and radium in people 41

4.4 Effective Kd 42

4.5 Clay soil profiles 42

4.6 Modelling the soil root compartment 42

4.7 Biosphere dose conversion factors 42

4.8 Understanding containment requirements 42

4.9 Effect of climate and environmental change on radium containment and mobilisation 43

APPENDIX A. LIST OF PARTICIPANTS 44

APPENDIX B. REFERENCES 46

1. INTRODUCTION

Radium-226 (Ra-226) is a member of the uranium-238 (U-238) series decay chain and is found naturally in minute quantities in a wide range of uranium-bearing rocks. Mining and processing of these rocks for a wide range of industrial purposes, gives rise to waste containing, in most cases, elevated concentrations of Ra-226 relative to those normally found in nature.

Radium-226 is of particular interest in safety assessments for the geological disposal of spent nuclear fuel, where in-growth of U-238 daughters may lead to additional input to the biosphere (the surface environment normally accessed and used by humans) over long timeframes. Radium-226 has a long half-life (1,600 years), grows in from even longer lived pre-cursors, and has a high radiotoxicity. It is relatively physically and biologically mobile. These factors combined mean that Ra-226 is of particular interest in risk assessments for the disposal of spent nuclear fuel and other radioactive wastes.

As a trace element, the behaviour of Ra-226 in the environment is complex and is affected by many biological and physicochemical factors. An interdisciplinary approach is therefore required in order to make appropriate predictions of long-term environmental impact of Ra-226, based on knowledge of the mechanisms determining radium mobility and behaviour both in and between different ecosystem components. As such, an international workshop was organised with the aim of bringing together organisations concerned with radioactive waste management, technical support organisations and research institutions to review the latest information on Ra-226 behaviour in the biosphere with a view to improving confidence in long-term safety assessments of public radiation exposure arising from waste management activities.

The workshop was held on 4-5 May 2010 and was hosted by Andra, Châtenay-Malabry, France. The workshop provided an open forum for presentation and discussion of the behaviour of Ra-226 in the biosphere. This report provides an overview of these presentations and discussions.

1.1 OBJECTIVES AND SCOPE OF THE WORKSHOP

The objectives of the workshop were to:

- provide an open forum for presentation and discussion of environmental processes governing radium behaviour in the biosphere with a view to improving confidence in prospective long-term safety assessments of public radiation exposure; and,
- develop recommendations for the direction of continuing research as input to long-term radiological assessment.

1.2 PARTICIPATION

The workshop was attended by 29 participants from 7 countries, representing a range of operators, regulators, researchers and technical support organisations. Participants are listed in Appendix A.

1.3 REPORT STRUCTURE

Section 2 provides an overview of each of the presentations made during the workshop and related discussions. The main gaps and uncertainties identified as a result of presentations and discussions are outlined in Section 3. Recommendations for further work to address remaining knowledge and data gaps relating to Ra-226 behaviour in the biosphere are presented in Section 4. References are given in Appendix B.

2. PRESENTATIONS

Provided below is an overview of each of the presentations given during the workshop. Points of discussion arising as a result of each presentation are also provided.

2.1 OVERVIEW OF DATA INTERESTS BASED ON EXAMPLE ASSESSMENTS FOR WASTE MANAGEMENT (GRAHAM SMITH)

Radioactive waste management organisations are interested in the long-term accumulation and cycling processes associated with Ra-226, following release to the biosphere from geological repositories. Ra-226 is not always present initially, but grows in from the decay of uranium that is present in the waste. In this case, the anticipated release of Ra-226 can be very delayed, well beyond 10,000 years, but then may continue for similarly extended times. For example, daughters of uranium radionuclides (including Ra-226) were among the principle radionuclides of interest identified in assessment of the generic deep geological disposal concept of the UK Nuclear Decommissioning Authority, with the peak doses occurring at time periods of around 1 million years post-closure[a]. Ra-226 was also a principle radionuclide in the EPRI assessment of the safety of disposal of Spent Fuel (SF) and High Level Waste (HLW) at Yucca Mountain.

Ra-226 is also of interest for near-surface disposal facilities, for example, it is one of the key radionuclides in the long-term at the UK Low Level Waste Repository (LLWR) due to the in-growth from uranium. Release from near-surface facilities would be almost directly into the biosphere; there are also more mechanisms for potential disruption to the emplaced waste, both natural and due to humans.

Other sources of long-term Ra-226 discharge to the biosphere include a variety of facilities for disposal of NORM (Naturally Occurring Radioactive Material) wastes, such as uranium and other mine and mill tailings, phosphogypsum piles and scale from the oil and gas industries. Over the years there have been a number of monitoring programmes at some of these sites and this may provide valuable empirical data on the migration of radium through soils and uptake by biota, including humans. (See for example, assessments made for the Olen site in Belgium, recorded in IAEA-BIOMASS-6).

Exposures from Ra-226 may include significant contributions via ingestion of contaminated crops and animal products, and drinking water; inhalation of dust and external irradiation from contaminated soils and sediments. The range in distribution coefficients (Kd) can be large[b] and this will greatly affect dose calculation results for all these pathways. There is a need therefore to understand the influence of different environmental conditions on Kd in order to guide appropriate parameter value selection for site-specific assessments. There is an additional need to be able to reliably estimate uptake into crops and hence into animals.

Ra-226 may be a significant source of exposure itself, but it is also of relevance as a source of exposure due to the in-growth of Rn-222, Pb-210 and Po-210 from radioactive decay. There is therefore a need to understand the environmental behaviour of radium to determine the potential exposure pathways arising from its progeny. This is also relevant to the uranium parents of Ra-226;

[a] NDA Report N-080

[b] Some of the Kd values reported in the literature may be determined by co-precipitation, e.g. with barium, rather than sorption.

any study on the behaviour of radium should therefore consider the entire decay chain to ensure that migration through the biosphere is accurately represented – each radionuclide within the chain will display different biogeochemical behaviour, thus potentially affecting the distribution of progeny in the biosphere.

For all the Ra-226 related wastes, and for all the methods of disposal, common dose assessment issues arise, including long-term decay and in-growth taking into account the effects of environmental change. Understanding the key factors affecting radium migration is vital for dose assessments.

Discussion

In discussion, it was noted that the IAEA Technical Reports Series No. 310 on the behaviour of radium in the environment is now more than 20 years old and a programme is in place to update the document. It is not intended that this will comprise an encyclopaedia-type approach, but rather that a handbook will be produced. An initial meeting of the TRS-310 working group is planned for July 2010 at which the content of the handbook will be identified. It is anticipated that the output of the BIOPROTA radium workshop will be used to inform discussions during this meeting.

2.2 OVERVIEW OF THE EFFECT OF PHOSPHOGYPSUM AMENDMENTS IN AGRICULTURAL SOILS (RAFAEL GARCIA-TENORIO)

Phosphogypsum is a by-product of the production of phosphoric acid, which itself is produced from phosphate-containing rocks. Radium present in the rocks (up to 1,000 Bq/kg) is largely retained in phosphogypsum, which leads to the in-growth of both Pb-210 and Po-210. Morocco has historically been a large producer of phosphogypsum; one of the worlds largest factories is located in Spain between the Odiel and Tinto Rivers. Annual production has been of the order of 3 million tonnes phosphogypsum.

Historically, a proportion of the phosphogypsum (up to 20%) was discharged to the Odiel River with the remainder being stored on land. However, in 1998 the practice of discharging phosphogypsum to the environment ceased and subsequently 100% has been stored on land. It is estimated that wastes of the order of 80 million tonnes now cover an area of 1,200 hectares of marshland. This area is within 500 m of the nearest human habitation.

Phosphogypsum has been used as an agricultural soil amendment to improve soil productivity. It is particularly useful in diminishing sodium saturation in saline marsh soils that have been reclaimed from the sea. The use of phosphogypsum in this manner is permitted under Spanish regulations – no mention is made in the regulations of radioactivity limits for phosphogypsum used for soil amendment. A three-year field study has therefore been conducted as to the radiological consequences of this agricultural practice. During years one and three, soil was treated with phosphogypsum at a rate of 25 tonnes per hectare and ploughed to a depth of 30 cm. samples were taken and radium measurements taken – higher values were recorded in the top 30 cm of soils compared with greater depths.

Since phosphogypsum is ploughed into the soils, dilution is substantial and no short-term radiological impact was noted:

- There was no statistical difference between treated and control sites, but this may relate to historical treatment of control sites with phosphogypsum;
- Soils were irrigated and drainage waters sampled – no differences were noted between treated and control sites;

- No radium was detected in the fruits of tomato plants grown on the soils. However, cadmium is also associated with phosphogypsum and has a similar behaviour to radium. Cadmium concentrations in tomato fruits were an order of magnitude higher on treated soils compared with controls.

To determine long-term impact, control plots (those with historical phosphogypsum treatment) were compared with plots outwith the reclaimed area (no historical treatment). No radium enhancement was noted in the upper 30 cm of plots outwith the reclaimed area whereas some enhancement was noted in the historically treated plots. The baseline concentration was 22 Bq/kg compared with 35 Bq/kg in treated soils. This indicates that there is some long-term, cumulative impact. However, dose consequences are minimal.

Radium activity concentrations in aerosols during application were also measured and a dose rate of 125 µSv/y was calculated on the basis of an 800 hour/y occupancy.

The dispersion of radium in the Odiel and Tinto Rivers resulting from historical releases was also studied. The rivers flow into a tidally-dominated estuary which serves to force material back into the river system rather than releasing to the marine environment. Studies have shown that radium is present at concentrations up to 225 Bq/kg.

Chronological archives are associated with sediment cores – a peak is noted that coincides with the period of discharge to the river (dates coincide with Cs-137 peaks monitored). The radium therefore appears to be immobile in sediments – no upward or downward migration was noted.

Soil to plant transfer studies are currently planned.

Discussion

The chemical form of radium appears to be a key factor in its environmental behaviour and the suggestion was made that radium mobility should be discussed in terms of the chemical form / compound. The soils in the test region are alkaline and this will affect radium mobility since mobility is related to pH. Alternative mechanisms for treating phosphorus-bearing rocks to derive phosphoric acid are likely to alter the chemical form and thus behaviour of radium in phosphogypsum.

The results of the research were noted to be similar to the results of modelling work conducted by ANDRA in which a high Kd is applied resulting in a low transfer factor. Modelling studies therefore suggest that radium is mainly an external irradiation hazard. The results were also noted to fit well with literature data.

It was also noted that there would be interest in looking at the differences in bioaccumulation between radium and polonium.

The phosphogypsum factory is due to close within a year after which time the area will be restored. It is currently planned that the area will be used for recreation activities (park / museum) and prolonged occupancy is therefore not anticipated, with perhaps the exception of museum and park employees. The accumulation of radon in buildings cannot be excluded.

Although phosphogypsum is used in some countries as a construction material (e.g. Brazil), this is not a practice employed in Spain so radiological impacts from external exposure to phosphogypsum-derived construction materials is not of concern.

2.3 REVIEW OF THE RADIOLOGICAL CHARACTERISTICS OF THE ENVIRONMENT AROUND A FORMER URANIUM MINE SITE IN FRANCE: THE CASE OF THE SAINT PIERRE SITE IN THE CANTAL DISTRICT OF FRANCE (MARIE-ODILE GALLERAND)

Uranium mining in France took place over a 50 year period from 1945 to 1995. During this time, 76,000 tonnes of uranium were produced. Radium-rich waste materials resulted from the physical and chemical processes involved in milling uranium ore. It is estimated that there were 50 million tonnes of tailings produced. Since mining ceased, around 200 sites have been remediated as required by French regulations. Some sites are still under statutory monitoring, including the Saint Pierre site in the Cantal district of France.

The Saint Pierre site consisted of a sedimentary ore deposit that was formed some 30 million years ago. The site is surrounded to the west, north and east by the Dordogne river. In south there is a man-made lake. The site was mined as an open pit and contributed around 2% of the entire French uranium produced. The site now consists of a large tailings and low ore area. Dredged material from the lake to the south is also contained at the site. Outside the mined area there are still natural uranium rich areas present. The site has two drainage areas; northerly drainage to the Dordogne and southerly drainage to the man-made lake.

In 2005, AREVA (the former site operator) undertook expert analysis of the site to produce a radiological data set, including details of the site hydrology, which has identified the discharge points to the north. Diffuse fluxes to the south nevertheless exist. Complementary investigations have been performed by IRSN.

The tailings deposits in the former settling basins have no near-field engineered barriers, for example, of the type used for a typical radioactive waste repository; around 3 Bq/l water is discharged from northern discharge point. A dam has been constructed to contain disposals in settling basins. The highest concentrations of uranium and radium have been measured in discharges from the base of dam. An on-site water reservoir has significant concentrations of uranium and insoluble radium. Sediments on the site are also contaminated; a former trench that collected mill water contains high concentrations of both uranium and radium. Radium concentrations of 70,000 Bq/kg have been recorded. Sediments rather than water are the main sink for radium contamination.

The south site drainage area has a number of drainage/recharge areas, which have been monitored. Uranium-rich materials at bottom of the manmade lake have been identified. It is evident that the tailings have contributed to the radioactivity in the south drainage area; however the un-mined mineral outcrop has also contributed.

A number of food-chain analyses have been undertaken around the site. Uranium-series radionuclide concentrations in fish from the on-site pool are significant at 60 Bq/kg radium compared with 0.6 Bq/kg in fish collected from the Dordogne River, even if the comparison is not appropriate (river versus water pool). No fish were sampled from the lake since they are absent as a result of dredging activities.

Saint Pierre kitchen garden produce (leek, potato and apple) are routinely analysed. Radium concentrations are enhanced compared with the control site for some produce; however many samples are below limits of detection for some uranium-series radionuclides. For example, polonium has not been detected, but the need to reduce the limit of detection has been identified to ensure measurements can be made if it is present, due to the high radiotoxicity of this radionuclide, particularly by the oral route. Radium concentrations in pears are enhanced; however, the tree is located in an area with significant gamma dose rate (200-400 nGy/h) and the pear concentrations are therefore directly linked to natural mineralisation in un-mined area soils. Traces of radium have been monitored in the milk of cattle grazing near the site. There is no detectable contamination in the Dordogne River or in fish.

A new surveillance programme is required to be undertaken to ensure all relevant sources of contamination that could give rise to farmer doses are included. At present, samples are collected on the basis of locality relative to the site rather than from consideration of transfer pathways.

Due to the uranium-rich deposits in the area, natural background has to be taken into account in order to perform a calculation of the additional dose.

Discussion

The benefit of producing a publication detailing the transfer factors arising from the monitoring programme was noted. However, there would be a need to fully review the sampling and monitoring procedures in order to determine such factors. The age of produce (e.g. pear tree) would need to be taken into account as would routes of uptake. For example, uptake of radium to the pear tree would be via soil. However, some in-growth could occur from parent radionuclides present due to the longevity of trees. Radon could be present as a result of radioactive decay of radium, but also from atmospheric uptake from radon released from soils.

Differences that were noted in activity concentrations between discharges from the north and south drainage areas can be partly linked to the dilution of the release. Fish analysis took account of different tissues. The lake has been dredged and it is not therefore possible to determine the historical timeline of releases from sediment profiles.

It was suggested that the overall impact radiological impact in the area of the site form uranium at the site may have been reduced in the long-term due to the removal of uranium during mining activities. The high natural background in the un-mined area compared with that in the mined area is indicative of this. However, it was noted that both physical and chemical changes have occurred on the site as a result of mining activities.

2.4 URANIUM AND ITS DECAY PRODUCTS IN THE HUMAN FOOD CHAIN (MARION JEAMBRUN)

Marion Jeambrun is a first year PhD student with Strasburg University, supported by IRSN and AREVA. The planned research programme was presented.

To date there have been few studies on natural radionuclides in French foodstuffs. This may in part be due to measurement difficulties such as the lack of equilibrium between decay products in the foods and the different uptake routes that are possible, which makes interpretation of results difficult. For example, there can be difficulties in determining whether the source of radioactivity is uptake from soil, irrigation water or atmosphere.

A review of the scientific literature has shown that soil to plant transfer has been analysed through both *in situ* and *in vitro* experiments. However, the contributions from different sources (irrigation water, atmospheric deposition and soil uptake) are seldom studied. With the exception of thorium, soil to plant transfer is greatest for leafy vegetables. Cereals generally have low transfer factors. Radium activity concentrations do not follow the U-238 results from uranium mining areas; there is a need therefore to study each element in its own right.

The aim of the research programme is, therefore, to determine the radiation baseline for uranium, thorium and their decay products in foods, with a focus on those foods associated with high consumption rates and the radionuclides with high dose conversion factors; i.e. those foods and radionuclides contributing most to human dose. Due to a general lack of data, Bi-210, Ac-227 and Pa-231 are also being studied. For foods that are stored for a significant period of time (e.g. wine), the content of daughter radionuclides will be analysed. The research programme also aims to look at the variability of activity concentrations in different regions of France, including atmospheric, soil and irrigation water concentrations.

Radiation doses to people in France from natural radioactivity are being investigated through literature research. The world-wide average annual effective dose from ingestion of uranium and thorium series radionuclides is 110 µSv/y, as estimated by UNSCEAR (2006).

Three groups of soils have been identified in France that can be distinguished on the basis of uranium activity concentrations. The origin of these groups of soils is being studied and one high (71 Bq U/kg), two intermediate (38 - 44 Bq U/kg) and one low activity (27 Bq U/kg) sites have been selected. The initial sampling campaign will begin in May and will look at the difference in transfer through the food chain at each site (e.g. cattle feed, milk, cheese). All foods will be washed in tap water prior to analysis as if they were to be consumed.

Laboratories undertaking analyses will undergo a robust inter-comparison exercise based on rosemary samples (with corresponding soil, irrigation water and atmospheric particle samples).

Discussion

Rosemary plants will be specifically analysed since they are irrigated with groundwater with known activity concentrations. Efforts will be made to separate plants from any surface dust, which would be analysed separately in order to try to determine the relative contributions to plant activity concentrations. It is not intended at this stage to analyse different parts of plants such as roots and shoots to determine distribution factors within plant types.

The importance of matched plant and soil samples for analysis was noted, in that it enables soil to plant transfer factors to be calculated. However, the scientific community would benefit greatly from the data generated if full soil analysis was also conducted as this enables users to select the most appropriate parameters on the basis of similarity in soil conditions.

2.5 RADIUM MOBILITY AND CYCLING IN RE-VEGETATED MINING DEBRIS (YVES THIRY)

Yves Thiry presented results obtained under a SCK.CEN project on a uranium mining-debris site in Germany. The majority of the results are available in the public literature (Thiry & Van Hees (2008) Science of the Total Environment, 232, pp 111-117; Thiry & Van Hees (2007) In Zhu, Y., Lepp, N. and Naidu, R. (Eds), ICOBTE 2007, 15-19 July 2007 Beijing, China, p. 449-450).

The remediation of uranium sites often involves phyto-stabilisation whereby areas are covered with topsoil and perennial vegetation such as forest trees established. This makes the system more durable by reducing water infiltration (rain water is intercepted by plants) and soil erosion. The aesthetic aspect is also improved.

Forest function involves the cycling of elements. There are two main capacities for forests with respect to elements – retaining capacity (storage with biomass accumulation) and recycling capacity (biological cycling with biomass turnover). Forests can absorb a large amount of calcium, an analogue for radium. It was therefore considered likely that radium would also be absorbed by forests. The study therefore looked at the cycling of radium between the soil, tree and forest floor systems and investigated the main properties governing the cycling of radium over a period of decades.

The study focused on a 35 year old pine stand growing on mining debris in Germany. The substrate on the mining heap is derived from a mixed rock source and is very stony – 70% of particles are greater than 2mm. The soils are alkaline and have a high calcium, magnesium, iron and aluminium content. Radium concentrations are of the order of 1100 Bq/kg.

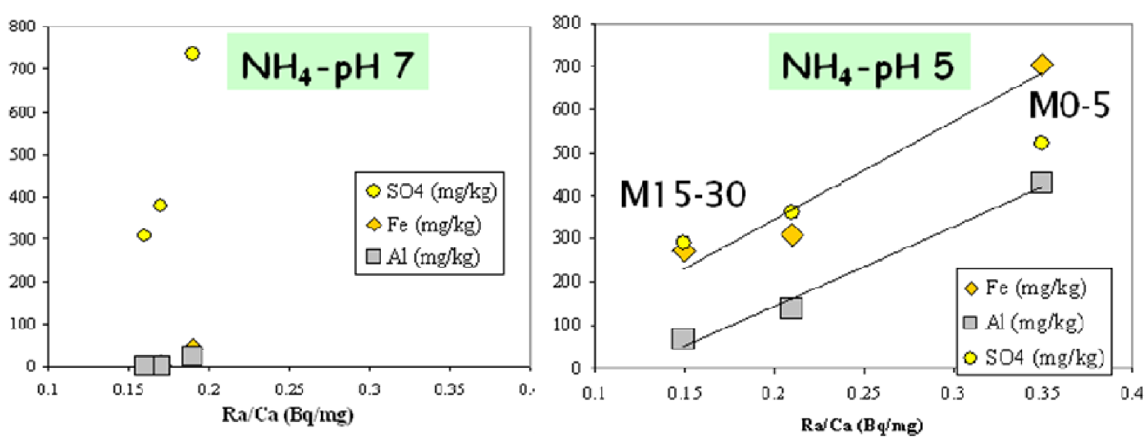
Annual fluxes were investigated. Need (the amount of an element required to produce new plant material) and uptake from soil were investigated and internal transfers calculated.

Calcium 'need' was calculated 29 kg/hectare (ha), uptake was 51 kg/ha, which equates to 0.8% of the ammonia-exchangeable calcium pool. An internal transfer of -17 kg/ha was calculated (the value was negative since the calcium was transferred to old rather than new tissues). Of the calcium taken up into the tree, 25% was retained and 75% recycled (returned to soil).

In the case of radium the 'need' was calculated as 20,000 Bq/ha and uptake was 24,000 Bq/ha. . The radium annual uptake represents a low fraction of ammonia available radium in soil (i.e. 0.002%) when compared with that for calcium. Internal transfer was -4,000 Bq/ha. Of the radium taken up into trees, a greater proportion was retained when compared with calcium; 40% was immobilised in the tree and 60% recycled. A radium budget for wholes trees showed that the majority of radium was allocated to the roots (88%) with a much lower proportion transferred to the above ground plant.. Fifty-four percent of radium in the below ground biomass was associated with fine roots. Roots therefore appear to be an important component of radium cycling and it may be important to represent them in a long-term assessment context.

In total, 0.0054% of soil radium was calculated to be currently retained by trees while 0.06% was taken up and accumulated in the forest floor. Assuming equilibrium conditions, the residence time of radium in forest floor is therefore greater than 200 years, which is inconsistent with other results. It is therefore postulated that, in early years, more radium was taken up by trees and is now locked up in humus.

Investigations of the soil pH gradient indicated that radium mobility is very much linked to decarbonation of surface mining debris. However, calcium and radium seem to have different chemical responses to acidification. Kd values were higher for radium than calcium, thus radium is less mobile than calcium in the soil profile studied. In the upper acidified layer, a specific adsorption pool for radium was attributed to the formation of sparingly soluble Fe-Al oxyhydroxides.



It was concluded that, at this site, pine vegetation development and biomass turnover had a negligible effect on radium cycling. However, roots may be of importance for long-term modelling and root production and decay may warrant further consideration. Acidification leading to de-carbonation of surface soils was found to be an important regulator of calcium and radium mobility. Organic matter and sulphate were not found to be retarding factors. Vegetation is expected to reduce hydrological losses of radium but, at a soil level, natural attenuation processes were not shown to promote soil remediation of radium.

Discussion

The association of radium with roots was believed to be with the living part of roots since roots were well washed prior to analysis. However, surface binding cannot be ruled out. Analysis of the uranium distribution in trees indicated an association with senescing organs, which is thought to be a detoxification mechanism.

2.6 RADIUM SORPTION ON SOILS (GRAHAM SMITH ON BEHALF OF ABDESSELAM ABDELOUAS)

The presentation focused on the key processes associated with radium sorption onto soils.

There is only one aqueous species for radium (Ra^{2+}); however various aqueous complexes are known (e.g. $RaOH^{+}$, $RaCl^{+}$, $RaCO_3$, and $RaSO_4$).

The WHO guidance level for Ra-226 in drinking water is 1.0 Bq/l whereas for Ra-228 it is 0.1 Bq/l.

Factors affecting radium speciation include pH and salinity (increased salinity leads to an increase in radium concentrations in groundwater). A good correlation has been noted between radium and total dissolved solids. There is a strong retention of radium in soils, particularly in alkaline conditions – co-precipitation may play a key role in soil retention. A strong link between radium and colloids has also been observed (although the size of colloids that the data refer to is uncertain and this will affect transportability). It is also possible that the retention of radium in soils will be affected by the concentration of calcium due to competition for soil sorption sites.

Discussion

In relation to the potential competition between radium and calcium for sorption sites it was noted that if calcium does compete with radium then the addition of calcium to soils should make radium more mobile. However this is in contradiction to other data. It was also noted that if competition did occur that this would also affect uptake by plants. However, it was argued that calcium would not be able to remove radium from soils due to the difference in electrical charge. It was therefore argued that there would be competition in relation to uptake but not sorption.

2.7 THE BEHAVIOUR OF RADIUM AND ITS PROGENY IN MAMMALS (MIKE THORNE)

There is on the whole a lot of information available on the behaviour of radium and its progeny in mammals, including kinetic data. This is largely due to mammalian laboratory data being the basis for developing human dosimetry standards and the need to interpret epidemiological data on cancers induced in humans due to exposure to Ra-224, Ra-226 and Ra-228.

Following inhalation, most forms of radium are moderately well retained (Type M) in the lungs, but Type F and S may be observed for some chemical forms. Lead is quite quickly removed from lungs (Type F) except when in the form of mineral dust (Type M and S). Polonium is moderately retained (Type M).

For gastrointestinal absorption there is a significant trend in absorption with age (as demonstrated by rat absorption data – 79% of radium was retained in weaning rats, 11% in juveniles and 3% in adults (Taylor et al., 1962)). Fasting was found to lead to an increased uptake (factor of 2) in older animals. Gastrointestinal absorption data are as follows:

- Humans: Drinking water and dietary – 0.15 to 0.21 (ICRP, 1973)[a]
- Dairy cows: 0.03 (Sansom and Garner, 1966)
- Sheep: > 0.01 (based on faecal excretion of 98-99%; Khademi and Mahdavi, 1974). However, radium has also been shown to be excreted in faeces following intravenous injection and these data should therefore be used with caution.

Pregnancy appears to affect absorption through the gastrointestinal tract as indicated by data on the effects of pregnancy on radium uptake in humans (Smith et al., 2003, based on Fell et al., 2001) whereby an increase from 0.2 in the first trimester to 0.4 in the second and third trimesters was recorded.

Data from paint ingestion studies indicate that dogs and humans show similar gastrointestinal absorption. Other mammals were 2 to 3 orders of magnitude lower.

Based on the available data, recommended gastrointestinal absorption factors for humans are:

- 0.05 to 0.3 for adults;
- 0.05 to 0.4 for 10-year-old children; and
- 0.08 to 0.8 for 3-month-old infants.

For lead, literature data on gastrointestinal absorption is largely available for the stable element. As with radium, enhancement of uptake has been observed following a period of fasting. This may result from an increase in calcium absorption following fasting with lead transferring with calcium. An excess of calcium can reduce lead uptake. Uptake by children is observed to be greater than that by adults (60% in infants compared with 20% in adults (ICRP, 1993)).

The physical and chemical form of lead can affect uptake and it has been suggested that high sulphur diets can lead to lead sulphide precipitation in ruminants which reduces gastrointestinal absorption (Underwood and Suttle, 1999).

In the case of polonium, the degree of absorption of dietary polonium (organic) differs from that of inorganic forms. Generally, gastrointestinal absorption in humans is around 50%, but lower values have been observed:

- Humans consuming reindeer meat: 0.3-0.5 (Hill, 1965; Kauranen and Miettinen, 1967; Landinskaya et al., 1973)
- Human consuming crab meat: 0.4 (Thomas, 1994)
- Human: 0.8 for some foodstuffs (Hunt and Allington, 1993)
- Male with myelogenous leukaemia: ~0.1 (Fink, 1950)

[a] References are provided in Annex 2

Similar data are observed for rats:

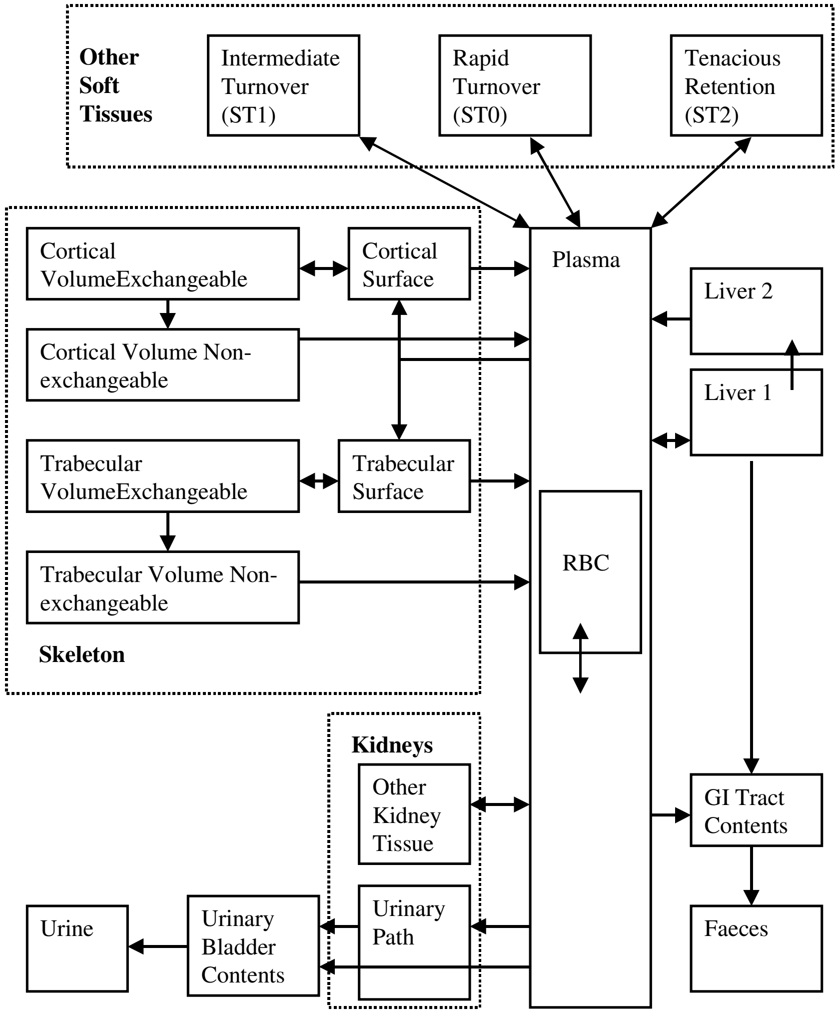
- Unspecified compound in rats: 0.03-0.06 (Anthony et al., 1956)
- Chloride in rats: 0.06 (Della Rosa et al., 1955)
- Rats ingesting Po in goats' milk: ~0.5 (McInroy et al., 1972)
- Rats ingesting Po incorporated in rat liver: 0.1-0.12

Systemic retention data for radium indicate high, but variable, bone to soft tissue concentration ratios (dry weight basis):

- Mule Deer: 4 (Mahon, 1982)
- American red squirrel and raven: 10 (Mahon, 1982)
- Rabbits: 100 to 10,000 (Holtzman et al., 1979)
- Cattle: 100 to 10,000 (Lapham et al., 1989)
- Cattle: ~400 (Linsalata et al., 1989)
- Pig: 4-40 (Nelson and Rust, 1967)

Sequestration of radium in skeleton relative to soft tissues is therefore variable. It is important to note that many of the earlier studies were conducted in a toxicological context, thus high concentrations were administered. Such concentrations result in skeletal damage and the data are not therefore representative of retention following more realistic uptake contexts. Similar whole body retention is observed between pigs and humans. Retention is not thought to change with age.

The ICRP recognises both exchangeable and non-exchangeable uptake of radium in the skeleton in its biokenetic model.



A similar compartmental approach is taken for lead, but the parameterisation differs. International consensus has not been achieved on the ICRP approach and alternative approaches have been proposed (e.g. Polig et al. (2004)). The ICRP model has been extended by Harrison et al. (2003) to include transfer to milk. Transfer factors for milk and beef are low (Morgan and Beetham, 1990):

- Skimmed milk of cows (chloride in drinking water): 0.00009-0.00022 d/L
- Skimmed milk of cows (contaminated hay): 0.00041-0.0007 d/L
- Milk of cows: 0.00013-0.00019 d/L
- Beef: 0.0005 (maximum 0.002), 0.00051-0.004, 0.00047-0.00086 d/kg

A retention co-efficient approach has also been used for radium. The approach considers the cumulative intake divided by the resultant body burden. The skeleton acts as lifetime accumulator for radium.

Radon-222 presents a more complex issue with regard to modelling behaviour in the mammalian system. Any model for radium in mammals, would need to consider accumulation in skeleton and decay, giving rise to Rn-222. The fraction of Rn-222 retained within the body will affect subsequent retention of Po-210. Radon-222 generated in soft tissues will escape from the body without decaying. However data suggest that radium is retained in bones with a difference in retention observed with time since intake due to incorporation into the mineral matrix of the skeleton. Retention within the mineral matrix would effectively prevent the release of any radon generated thus enabling decay to lead and polonium. The fraction of Rn-222 retained in mineral bone varies from 0.06 at 1 day to 0.4 at 27 years (ICRP, 1979).

Polonium is deposited primarily in soft tissues with the reticuloendothelial system having a greater retention, by around a factor of 10, compared with other soft tissues. Significant skeletal retention has been observed, but this is mainly in bone marrow rather than mineral bone (Naylor et al., 1991).

There are few data on transfer factors for Polonium largely due to the significant degree of in-growth from Pb-210. Those available include:

- Cattle: 0.00034 d/L (milk) (IAEA, 1994)
- Cattle: 0.005 (0.0006-0.005) d/kg (beef) (IAEA, 1994)
- Poultry: 2.3 d/kg (flesh), 7.0 d/kg (eggs) (Staven et al., 2003). Data based on review of similar elements and do not therefore directly relate to polonium.
- Value for human milk of ~0.029 d/L (Watters and McInroy, 1969)

Based on the review of available data for radium and its progeny in mammals it was concluded that there is sufficient data available to enable biokinetic models to be developed for Ra-226, Pb-210 and Po-210. There is also a reasonable basis for estimating losses of radon from soft tissue and mineral bone.

Discussion

There is a general preference for dynamic modelling rather than use of simple transfer factors in relation to the uranium-series radionuclides and it was recommended that kinetic models are run under different intake rates for comparison against literature transfer factors. Such an approach would help to uncouple uncertainties and allow underlying causes for differences to be attributed, e.g. links between the uptake of radium and calcium in diet.

For the loss of radon from the body, diffusive transport from soft tissues to the blood stream enables transport to the lungs and subsequent loss from the body during respiration.

2.8 ESTIMATION OF SOIL TO PLANT TRANSFER FACTOR OF RADIUM-226 (KEIKO TAGAMI)

A number of methods are available for estimating soil to plant transfer factors. These can be direct methods (radiotracer laboratory studies or field observations) or indirect methods (e.g. target element concentrations in soil and plant or use of multi-regression analysis for major properties of soil and plant samples).

In Japan, data have been gathered to support mathematical models for deep radioactive waste disposal sites. Due to the nature of such models, there is a need for robust data, with a preference for field data. Short-term radiotracer experiments are not so applicable. However, low Ra-226 concentrations in crops from field studies make direct measurements difficult.

Comparison of Japanese derived data and those within the IAEA Report TRS-472 shows a number of inconsistencies in that concentration ratios within TRS-472 are often much higher for many crops, particularly cereals:

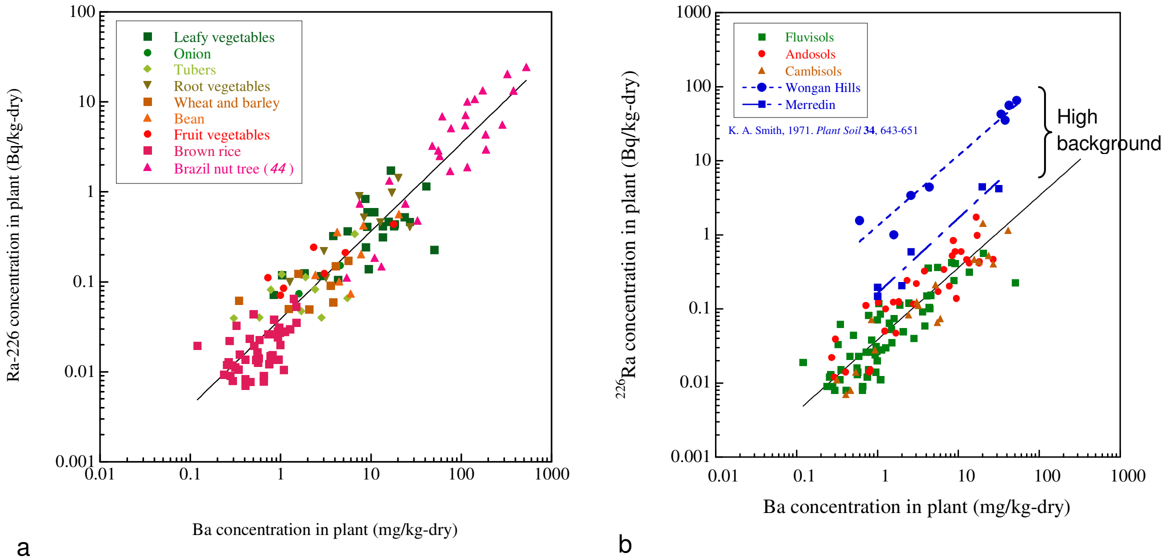
	n	GM/AM	GM/ TRS-472	Min.	Max.
Cereals (excl. rice)	8	2.7E-3	0.16	1.4E-3	4.9E-3
Brown rice	46	4.6E-4		7.1E-5	1.6E-3
Brown rice (Japan, Sasaki et al.)	2	4.7E-3			
Brown rice (China, Shang et al.)		4.1E-3 (Clay), 1.4E-3 (Clay),1.2E-2 (Loam)			
TRS-472, cereals (excl. rice)	24	1.7E-2		8.0E-5	6.7E-1
Leafy vegetables	24	1.1E-2	0.13	1.3E-3	5.8E-2
TRS-472 (Leafy vegetables)	77	9.1E-2		1.8E-3	1.3E+2
Fruit vegetables	7	4.2E-3	0.25	2.0E-3	1.5E-2
TRS-472 (Non-leafy vegetables)	44	1.7E-2		2.4E-4	6.3E+0
Leguminous vegetables (Seeds)	7	6.6E-3	0.47	3.4E-3	1.5E-2
TRS-472 (Seeds and pods)	40	1.4E-2		3.2E-4	6.2E+0
Root crops	11	9.3E-3	0.13	2.0E-3	3.3E-2
TRS-472 (Root crops)	60	7.0E-2		2.0E-3	5.6E+1
Tubers	10	2.2E-3	0.20	1.1E-3	1.0E-2
TRS-472 (Tubers)	45	1.1E-2		2.4E-4	3.9E+0

The objective of the study was to estimate concentrations in associated soils and plants. In total, 134 crops and corresponding soils were collected throughout Japan from agricultural fields. Samples were analysed for 58 elements. Crop samples were subject to radiochemical separation procedures prior to low-background liquid scintillation counting in order to detect Ra-226. Soil samples were measured for Ra-226 by Ge detecting systems. Soil samples were also analysed for water content, pH, soil particle size, major and trace elements (by ICP-OES and ICP-MS), including exchangeable calcium.

A good correlation was observed between uranium and radium concentrations in Japanese soils (Log(Ra)soil = 1.150 + 0.876 Log(U)soil) and world data (Log(Ra)soil = 1.102+0.934 Log(U)soil), giving an overall correlation of Log(Ra)soil = 1.11 + 0.934 Log(U)soil. It is therefore possible to estimate radium concentrations from measurements of uranium in soils.

In addition to uranium, radium was also found to be highly correlated in soils with thorium, beryllium and thallium.

In crops, radium was observed to be highly correlated with barium ($R = 0.87$ ($p < 0.001$), Figure a), but also calcium ($R = 0.83$ ($p < 0.001$)) and strontium ($R = 0.86$ ($p < 0.001$)). The correlation with barium was particularly important since a correlation was also evident between Ra and Ba in soils with different background activity concentrations. A different degree of bioavailability was observed in high background areas (Figure b), but where the Ra/Ba ratio is within the normal range, bioavailability would be expected to be comparable with world data.



a b

When transfer factors for radium were estimated on the basis of barium concentrations in crops and uranium concentrations in soils, a good correlation was observed between estimated and measured data. Although the correlation was strong, there is not a large amount of measured data for these elements. The study therefore looked at correlations between radium and major soil and plant parameter data, which are more readily available. In the case of soils, the following parameters were investigated:

- clay content;
- pH;
- cation exchange capacity (CEC);
- exchangeable Ca;
- exchangeable potassium;
- active Fe;
- water content;
- total carbon content (TC); and
- total nitrogen content (TN).

Correlations between radium and the elements P, Ca, Fe, K, Mg, Mn, Na, Cu and Zn were investigated for plants.

The greatest correlations were observed between radium and exchangeable potassium in soils (R = 0.5) and radium and calcium in crops (R = 0.8).

A multiple regression analysis was performed to identify those parameters that are most closely linked. Nitrogen and carbon were found to be closely correlated and, as such, nitrogen was removed from further analysis to ensure that the variables analysed were independent of one another. Subsequently, measured and estimated values were compared and a good correlation observed. However, the ability to apply this approach to other countries is uncertain.

Overall the multiple regression approach gave the best results, but the initial approach of estimating radium concentrations in soils and crops individually on the basis of uranium and barium concentrations may be more widely applied.

Discussion

In discussion the benefits of this approach were discussed. Due to the long-term modelling required for radioactive waste disposal, there is uncertainty around the soil conditions that will be present in the future. This approach could therefore be applied as a means of undertaking uncertainty analyses on the basis of the range of possible variations in soil parameters for relevant soils.

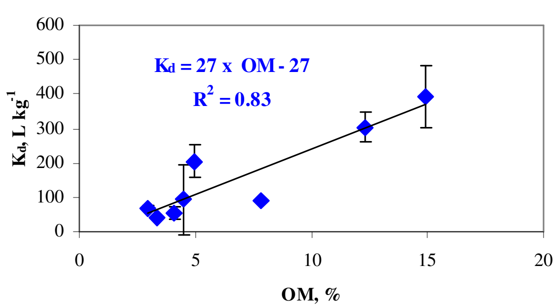
It was noted that there may be benefit in comparing the Japanese data with the stable element data for Canada produced by Steve Sheppard. It was considered that similarities would be observed for soil samples, but that, due to the differences in growing conditions, differences are likely to be observed for plant data.

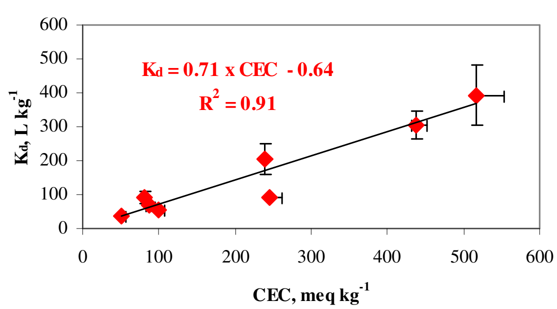
2.9 PREDICTING RADIUM AVAILABILITY AND PLANT UPTAKE: PROPOSAL FOR NEW BEST ESTIMATES OF KD AND TF FOR RADIUM (HILDEGARDE VANDENHOVE)

Many reported Kd and TF values are determined without any recording of the underlying factors such as soil parameters. There is therefore limited information linking radium bioavailability to soil parameters upon which changes in bioavailability can be predicted in relation to changing environmental conditions. The presentation detailed results from previous studies and also presented the results from the IAEA TRS-364 revision working group relating to radium in soils and uptake into plants (IAEA TRS-472, 2010; Vandenhove et al., 2009a,b).

Radium is strongly retained by soils and a long-term study has indicated that over a 9 year period following surface contamination, the majority of radium was retained in the top 15 cm of soil, giving a migration rate of 0.4 cm/y. The IAEA (1990) stated that the retention is controlled by availability of sorption sites on clay minerals and organic matter, with organic matter being ten times more efficient at retaining radium than clay. The relationship between Kd and organic matter content and other soil properties such as CEC was therefore investigated.

Results indicated a good linear relationship between Kd and organic matter content and Kd and CEC. Where these soil parameters are known it may, therefore, be possible to apply linear relationships to determine radium retention.





Radium retention correlations indicated that pH and radium mobility are not closely linked.

In order to investigate these relationships further, the IAEA TRS-364 revision working group established a set of data acceptance criteria to screen literature data in order to determine best estimate values for radium Kd in four soil categories (organic, clay, sand and loam). As a result of the acceptance criteria applied, only 8 references were retained, which gave rise to 51 data points, greater than in previous studies (IAEA, 2010).

The highest Kd was observed for clay soils, but no significant difference was observed between the different soil types due to the large spread of data:

Soil group	n	GM	GSD	AM/Value	SD	min	max	# ref
All Soils	51	2500	13	34000	130000	12	950000	8
All soils*	47	1800	10	11000	21000	12	100000	7
Sand	20	3100	8	9600	12000	49	40000	4
Loam	19	1100	17	15000	32000	12	120000	5
Loam*	17	710	14	8600	20000	12	80000	4
Clay	6	38000	12	200000	37000	696	950000	3
Clay*	4	13000	10	41000	47000	696	100000	2
Organic	1			200				1
Unspecified	4	1200	1	1300	500	785	1890	1

* excludes those experimental data that have extremely low Ca+2 content (much lower than that encountered in soil pore water).

This large range, combined with a lack of data for organic soils makes the application of the organic matter linear relationship approach difficult.

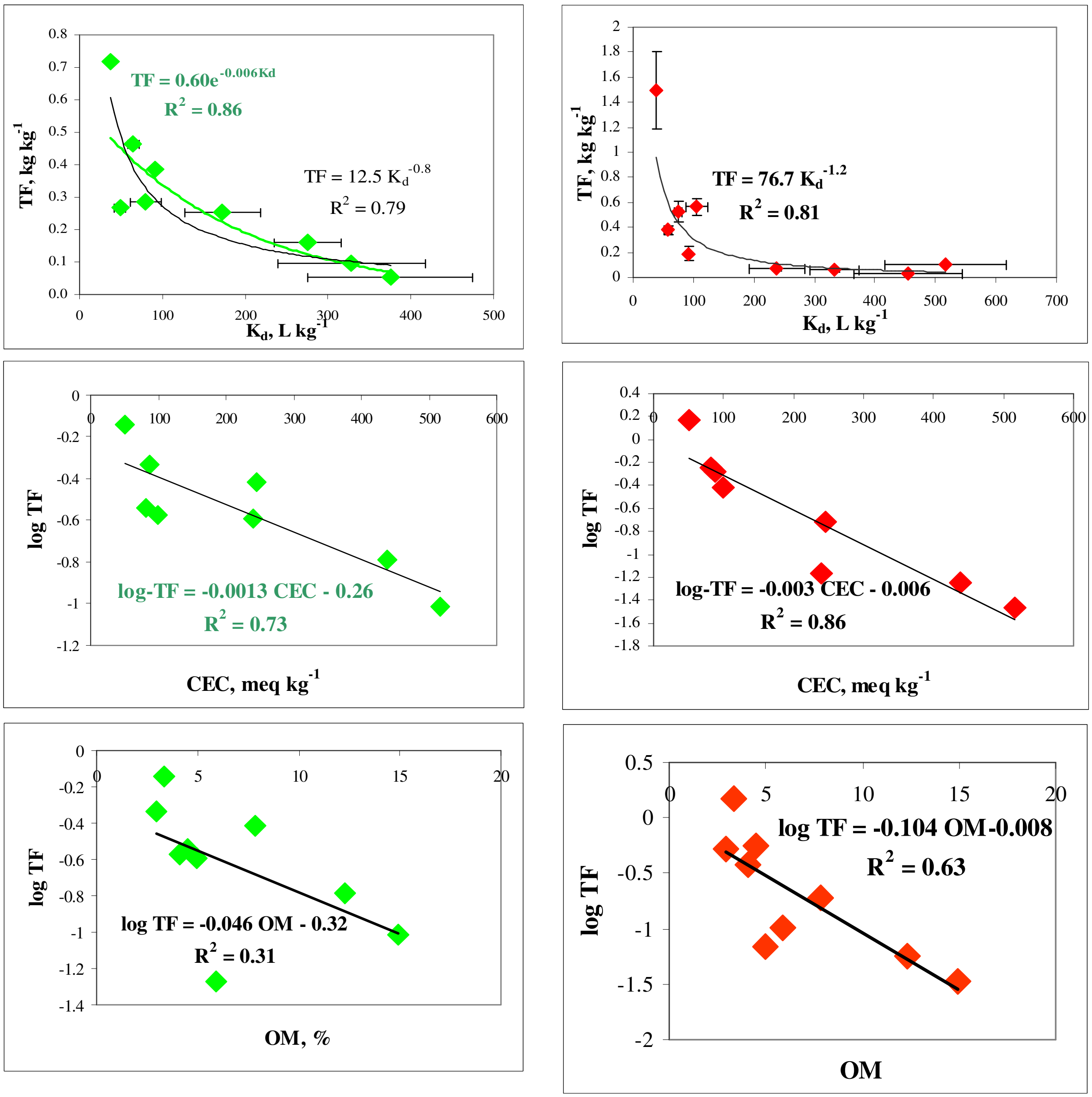
Below are presented the IAEA TRS-364 (1994) best estimates for Kd values in soils. An important discrepancy with the new best estimates of IAEA TRS-472 is observed for most soil types.

- Sand – 490
- Loam – 36,000
- Clay – 9,000
- Organic – 2,400

In contrast, Sheppard et al (2006) gives a single Kd for all soils of 47, which is substantially lower than other Kd estimates. The need to establish reasons for these differences was identified.

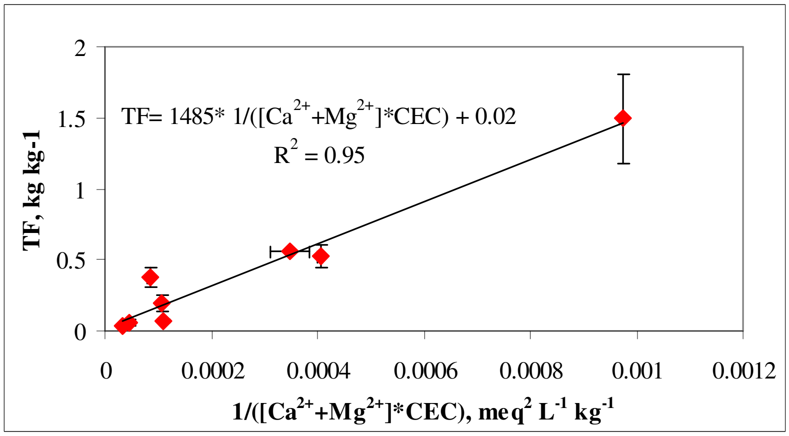
Calcium concentrations in soils appear to be linked to Kd and it was postulated that, where high Kd values are observed, there is competition between radium and calcium for sorption sites. Soil texture, however, was not strongly linked to Kd so soil texture classes may not be an appropriate way of classifying Kd.

In the case of soil to plant transfer, a significant negative correlation is observed between pH and radium uptake into clover. Such a relation was not observed in ryegrass, however. Similarly, in one study, a negative relationship was observed between radium uptake and silt and clay content of soils, but this was not the case in another study. A strong relationship is observed between soil to plant transfer factor and Kd, CEC and organic matter in both ryegrass (green) and clover (red). However, another study found no relationship between CEC and OM and uptake into plants. Such differences may relate to the particular characteristics of the plants and soils studied.

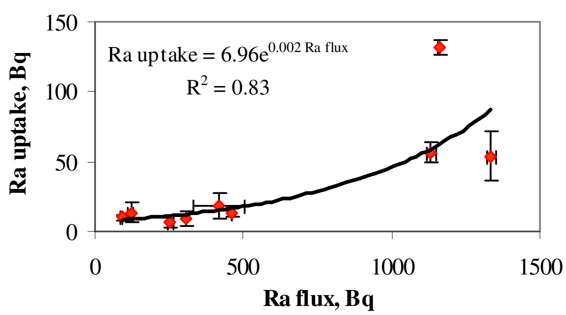


Based on the correlation between Kd and soil to plant transfer factor it is evident that, if radium is available in soil solution, it is available for plant uptake. This is not the case for other radionuclides such as uranium.

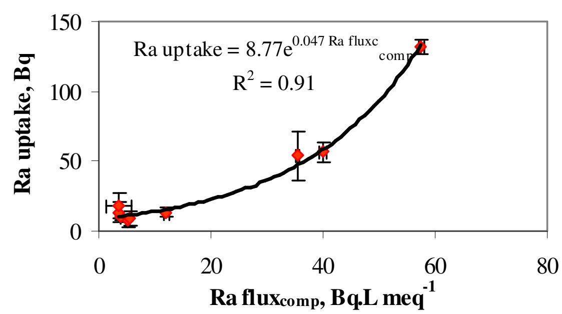
Mixed results are obtained when considering the correlation between radium uptake into plants and soil exchangeable calcium content. However, a very strong negative relationship is evident between the presence of divalent cations in soils and uptake of radium into plants. Divalent cations act as an inhibitor of radium uptake. CEC is observed to decrease uptake. Where both these parameters are combined there is a strong link between the competitive action between calcium and magnesium and the uptake of radium into plants.



Total radium uptake into plants and radium flux were also compared and a strong relationship was again observed (figure a). If competition is taken into account (i.e. the presence of cations) a stronger relationship is observed (figure b).



a) Ra-flux = [Ra] x water flow



b) Ra-flux$_{comp}$ = [Ra] x water flow/[M^{+2}]

Consideration was also given to the evidence for barium and strontium as tracers for radium. Available data suggest that strontium is not closely linked to radium, but for barium a stronger relationship is generally observed. However, in the case of loamy soils, which may have a naturally high barium concentration, the relationship is not so strong. Background concentrations of barium must therefore be considered if applying such a relationship to derive radium uptake by plants

Results concerning the relationship between plant calcium content and radium transfer are mixed and it appears that the relationship may be dependant upon plant type.

For the IAEA TRS-364 revision (TRS-472), a range of data acceptance criteria were employed to determine transfer factor best estimates. In total, 47 references met these acceptance criteria, giving a total of around 560 data points. Plants were grouped into nine major crop groups and, for each group, plant compartments were established. The study found no relationship between transfer factors for radium and soil concentrations. Leafy vegetables, root crops, herbs and pasture/fodder were found to have similar radium transfer factors. Non-leafy vegetables, cereals, legumes, tubers and fruit were again similar, but around an order of magnitude lower than for the leafy vegetables group. Overall the transfer factors derived were lower than for the previous TRS-364.

Consideration was given to global factors for soil categories (i.e. all plants within each of the soil categories of clay, loam, sand and organic). Transfer factors for sand and loam were generally slightly lower than for clay and organic soils. The GM between different soils is similar, but the range within soil type is large, which is an argument against categorising radium soil-to-plant transfer parameters based on texture classes.

No overall effect of soil properties on radium uptake into plants was observed when all data were considered together. However, some specific factors significantly explained the variation observed when specific crop types were considered:

- Negative dependency of TF on OM
    - Legumes: R^2= 0.42
    - Leguminous fodder: R^2=0.62
    - Natural pastures: R^2= 0.27
- Negative dependency of TF on pH
    - Leguminous fodder: R^2=0.48
    - Grasses: R^2= 0.32

No relationship was observed between radium uptake into plants and the presence of calcium, magnesium or potassium. Similarly, no effect of climate type was observed (data was available for both maize and potatoes to study this parameter). A difference was observed in transfer factors for Ra-228 and Ra-226. Overall the Ra-228 transfer factor was around 2.5 times greater than for Ra-226. Steve Sheppard has also determined a difference in plant transfer factors between these two isotopes, but there is no obvious explanation for the differences observed.

Discussion

The experimental data presented in relation to soils and soil to plant uptake experiments outwith the TRS-364 data compilations were standard soils collected from throughout Belgium. Soils with the most contrasting soil properties were selected for the study.

It was discussed as to whether other soil / plant properties could be important in determining radium behaviour. However, multi-attribute analysis had been undertaken and the parameters presented were responsible for the majority of the variation observed according to the results of the analysis.

There is a need to consider additional pathways of uptake into plants when looking at field conditions. Atmospheric uptake, including particle deposition, may occur and contribute to plant concentrations. Where such pathways exist, soil to plant transfer factors may underestimate plant concentrations.

The benefit of being able to access the database of soil Kd and plant uptake data was raised. Access would enable assessors to select the most appropriate data and, where any high values are identified, judgement could be made as to their applicability to the site of interest.

Finally, the need to ensure that soil characteristics are determined in studies relating to radionuclide uptake and environmental transfer was highlighted. Such data are invaluable for enabling data to be accurately interpreted and for results to be placed in context.

2.10 ENVIRONMENTAL BEHAVIOUR OF RADIUM-226 IN AQUATIC AND TERRESTRIAL ECOSYSTEMS (RACHID EL MRABET)

The National Centre for Energy, Sciences and Nuclear Techniques (CNESTEN) in Morocco is comprised of a large complex with a varied remit ranging from radiopharmaceutical production to radioactive waste treatment and disposal.

There were three aspects to the presentation.

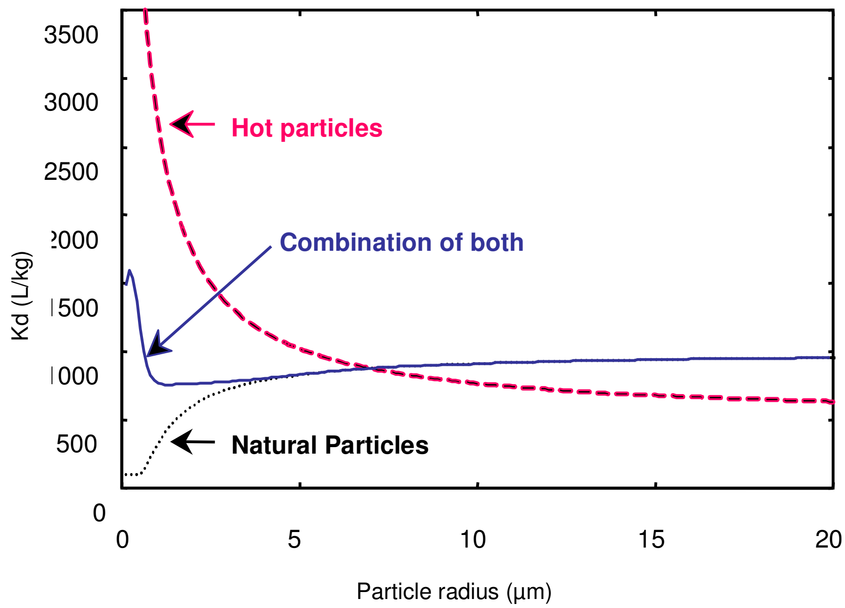
The initial focus was on the evaluation of impact of radioactive emulsion wastes associated with the phosphogypsum industry in aquatic systems.

Analytical approaches enable distinction of industrially derived radium from natural radioactivity on the basis of the location of radioactivity associated with sediment particles. Where radioactivity is naturally derived, the inner core of the sediment particles will contain radium. Surface contamination can result from radioactivity present in the dissolved phase as this absorbs to the particle surface. In contrast, industrial particles or 'hot' particles originating from industrial debris are generally of a much smaller size and can be distinguished from natural and/or surface contaminated particles.

A large variation (up to several orders of magnitude) has been observed in Kd, with variation being observed at the same site over time. Various physio-chemical factors can contribute, including particle size. Equilibrium Kd is defined as the ratio of the specific activity of the radionuclide in suspended matter (in Bq/kg) and its corresponding specific activity in the dissolved phase (in Bq/l). both surface and inner sediment activity concentrations should be taken into account in determining Kd.

Mobile ions can interchange with the µm-thick surface layer of particles. Where only man-made radionuclides are present, surface contamination rather than inner contamination will be present. In such instances, Kd will decrease with particle size. However, for 'hot' particles, inner contamination will be present. Where the radionuclide is highly soluble, such as is the case for radium, the particle surface would be expected to have a lower activity concentration than the inner particle, thus Kd would increase with particle size. For highly insoluble radionuclides, binding is largely in the surface layer with a lower concentration in the inner particle. In such instances, the contribution to Kd of the inner particle increases with particle size, while the relative contribution produced by the surface layer decreases.

Kd / particle size relationship curves vary depending upon the ratio between 'hot' particles and naturally occurring radionuclides. When curves for naturally occurring radionuclides and 'hot' particles are combined, a small peak is observed, the amplitude of which relates to the number of 'hot' particles present in the sediment sample.



The above technique has been applied to sediments in the Tinto River, Spain, which has historical contamination from the phosphogypsum industry.

Sediment samples were obtained from different regions of the river, including around the original discharge point. Sample particle size was distinguished and small particles, characterised as 'hot' particles were observed.

Both thorium and radium were measured. In samples collected close to the original discharge point, high radium activity was observed (3,300 Bq/kg) in the smallest sediment sub-sample (particle size 0.02 to 0.05 mm). Activity was found to vary with the sediment size fraction (970 Bq/kg (0.05 – 0.1 mm), 1,160 Bq/kg (0.1 – 0.5 mm), and 1,810 Bq/kg (0.5 – 2 mm)). Water activity concentrations however were constant at 40-80 mBq/l radium. The peak observed near the discharge point associated with small particle size is a strong indication of the presence of 'hot' particles. The measured Kd at this sampling site was much higher than at downstream sampling sites. Sampling site 2, downstream of the discharge point, had a lower Kd value as a result of additional input of sedimentary material which served to dilute the 'hot' particle contribution. The peak in activity concentrations with small particle size decreased with distance downstream from the discharge point.

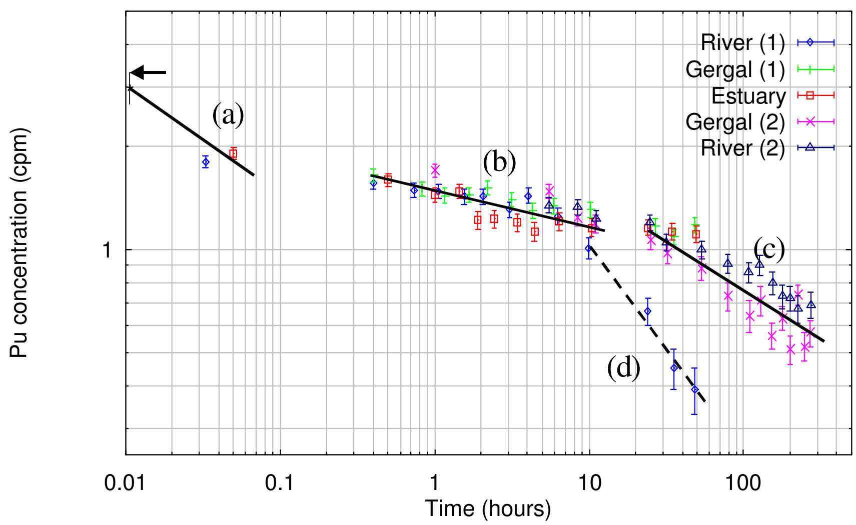
Since 'hot' particles are of a small size, they can be transported over large distances. No 'hot' particles were identified in the Odiel River. However, such particles were present at the confluence of the Tinto and Odiel Rivers.

The second part of the presentation focused on kinetic reactions between water and suspended matter using barium-133, americium-241 and plutonium-239 as tracers.

Suspended sediment can contain more radioactivity per gram that is present in 1 m^3 of water. Suspended sediment is therefore important in relation to the transport of radioactivity in aquatic systems.

One litre aliquots of natural water containing suspended sediment were transferred to glass beakers and spiked with a radiotracer. Stirring was conducted continuously. Over time, aliquots were taken and suspended sediment removed. Supernatant was then acidified and radioactivity concentrations measured. The behaviour of the radiotracer in relation to suspended matter could therefore be determined over time.

In the case of Pu-239 an initial fast absorption phase was observed, which was followed by a slower phase which occurred over a period of hours. A third very slow phase (over a 100 hour period) of absorption then followed.



The different sorption phases are explained by the accessibility of particle absorption sites to radionuclides. Particles are of an irregular shape. The initial fast phase corresponds to sorption of radionuclides on the readily accessible sites. The second, slower phase is associated with sorption onto less accessible sites. The third very slow phase relates to the migration of activity into the particle itself.

Three process steps are therefore evident and it was thus argued that a simple 2-box equation is not appropriate for modelling the sorption process. A good correlation between observed and calculated concentrations is obtained when a three-component equation is applied.

As a result of this research, it was concluded that variation in Kd results from differences in the specific surface area of the particulate material. It was also suggested that injection of particulate matter into water bodies could be used as a mechanism for decontaminating water.

Part three of the presentation focused on a radiological impact assessment for phosphogypsum wastes. Historically such wastes were discharged to the environment, but policy is now changing. Alternative approaches to disposing of phosphogypsum waste are therefore required, since the stability of phosphogypsum mounds is not adequate to permit long-term storage.

In some countries, use in agriculture or in road construction is permitted. However there is a potential radiological issue, largely associated with the emanation of radon. In the USA, a limit of 370 Bq/kg Ra-226 is in place for the use of phosphogypsum in agriculture. Such a limit is not in place in Spain however. This may be due to differences in agricultural practise. In the USA, surface application is the norm whereas in Spain the phosphogypsum is ploughed into soils thus increasing dilution. The practise of applying phosphogypsum helps improve and restore soils for agricultural use. An alternative use for phosphogypsum is in road construction. Such use can be associated with economic benefits by reducing the cost of road construction materials.

A dose assessment was conducted to determine the potential radiological impact of the application of phosphogypsum to agricultural soils. The average radium concentration in phosphogypsum in Morocco is 510 Bq/kg. Phosphogypsum was applied to an experimental farm and the effect of ploughing to different depths investigated. No uranium or thorium was detected in plants grown on the experimental site. Only a slight increase (around 3 Bq/kg) was observed in radium concentrations in soil when phosphogypsum was ploughed to a depth of 30 cm. Drainage waters were not affected in the short term.

The dose assessment considered uptake into crabs using recommended concentration ratios for molluscs and subsequent consumption by humans. The resultant dose estimate was less than 5% of the recommended limit.

Discussion

The need for defining the process applied for filtering sediment from water was highlighted with regard to derivation of Kd, with different processes having a large effect on resultant Kd. In this instance, a 0.045 µm filter pore diameter was applied.

Although phosphogypsum amendment of agricultural soils does not pose a radiological impact (on the basis of the dose assessment results presented), it was noted that chemical toxicity may result due to the presence of heavy metals including arsenic and cadmium. The addition rate to soils is very low so groundwater contamination risks are considered to be low. Uptake into plants would be dependant upon the plant type.

Consideration is required as to the economics of phosphogypsum amendment. Where transport over large distances would be required for agricultural application, alternative uses would be preferable on economic grounds.

2.11 DATA REQUIREMENTS TO DETERMINE EFFECTIVE DISTRIBUTION COEFFICIENTS FOR RADIUM IN SOILS (LARA DURO)

Previous studies have been undertaken to estimate effective Kd values (based on geochemical processes associated with the geosphere and Quaternary sediments) for a range of radionuclides by quantitative coupled groundwater flow and reactive solute transport simulations. The simulations hypothesised an injection of radionuclides from a geological repository at Forsmark, Sweden. Radionuclides were assumed to have passed through the geosphere by transport in deep groundwater, which flows along fractures in deformation zones. Shallow groundwater then flows toward surface water bodies (lakes, rivers etc). Different redox behaviour and retention characteristics were considered in relation to their effects on transport.

Uranium is associated with the precipitation of pure solid phases and sorption onto solid surfaces, which is dependent upon site conditions such as groundwater characteristics and pH. Uranium sorbs until a maximum concentration is reached, but then desorbs as a result of competition with other elements for sorption sites.

Radium and barium would be expected to co-precipitate. However, radium does not hydrolyse until pH 13 so sorption to clay surfaces is unlikely. Cation exchange was therefore assumed and the cation exchange rate for barium was applied to radium.

For the elements of interest the key processes were identified and consideration given as to whether appropriate thermodynamic data was available. Key processes and data availability are summarised below.

Retention process	^{14}C	^{129}I	^{36}Cl	^{94}Nb	^{59}Ni	^{93}Mo	^{79}Se	^{99}Tc	^{230}Th	^{235}U	^{135}Cs	^{90}Sr	^{226}Ra
Sorption onto organic matter	■	□	□	□	□	▒	□	▒	□	□	■	■	■
Sorption onto Fe-Mn-Al oxyhydroxides	■	▒	■	□	□ ✓	▒	▒ ✓	■	▒	□ ✓	■	■	■
Sorption onto phyllosilicates	■	■	■	■	□ ✓	■	■	■	▒ ✓	▒ ✓	□ ✓	□ ✓	□
Precipitation as pure phases	□ ✓	■	■	□ ✓	▒ ✓	▒ ✓	□ ✓	□ ✓	▒ ✓	▒ ✓	■	■	■
Association with sulfides	■	■	■	■	□	□	□	■	■	■	■	■	■
Association with carbonates	■	■	■	■	▒	■	▒	■	■	■	■	□ ✓	■
Incorporation into bacteria	■	■	■	■	■	■	▒	■	■	■	■	■	■
Association with phosphates	■	■	■	■	■	■	■	■	■	▒	■	■	■
Association with sulfates	■	■	■	■	■	■	■	■	■	■	■	■	□ ✓

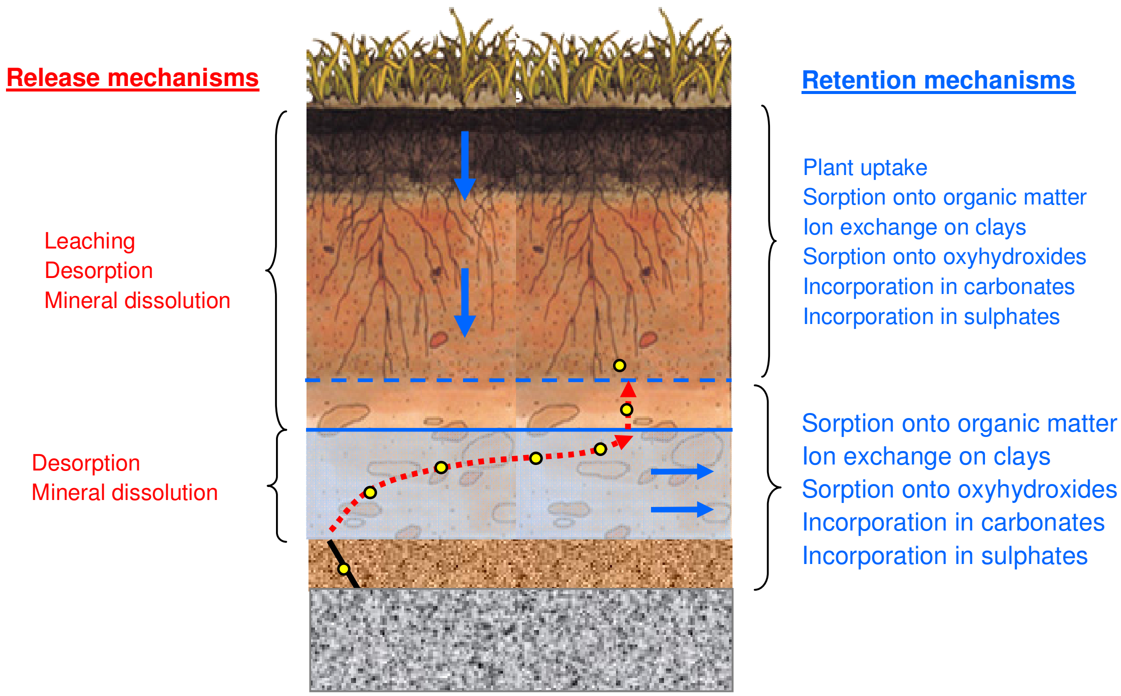
□ Processes likely to be active in the QD of Forsmark
▒ Processes not likely to be active in the QD of Forsmark
■ Retention processes not relevant for the indicated element
✓ Available thermodynamic data

Kd values will vary over time due to changes in many factors, including reduction in the concentration of a radionuclide as it migrates from the repository toward the biosphere. Effective Kd is therefore calculated on the basis of retardation factors over a 30,000 year simulation period. In most instances, the calculated effective Kd was lower than literature Kd values derived from laboratory experiments.

Once radionuclides pass through Quaternary sediments they enter the soil environment. Interest is then focused on what happens to the radionuclides in soils and calculating effective Kd for this ecosystem compartment.

In the case of radium, concentrations in the biosphere will depend upon how rapidly both uranium and radium are transported through the soil. In deep sea sediments, radium and uranium are enriched, whereas depletion is observed in oceans and lakes due to low thorium solubility and enhanced deposition.

Conceptual models are currently being developed for soils for which the identification of retention and release mechanisms is required, including the interaction of radionuclides with organic matter, which is a process not considered previously in effective Kd simulations for the geosphere and Quaternary deposits. Mechanisms included are demonstrated in the following diagram. Based on literature data, uranium is expected to be primarily associated with organic matter, whereas radium would be associated with carbonates and oxides.



Data are required for the parameterisation of transport models. The models required are particularly complex due to the need to consider reactive transport simulations in parallel with radioactive decay over time (i.e. Ra-226 cannot be modelled independently of U-238 and Th-230). However it is possible to couple the results from effective Kd model simulations with reactive decay.

The biogeochemical properties and parameters required for each soil horizon include:

- Percentage of each fraction (air, water, inorganic, organic) and sub-fraction (e.g. for organic → humus, roots)
- Mineralogical composition of inorganic fraction
- Particle-size distribution
- Type of humus
- Type and depth of roots
- Soil pH
- Soil temperature

Particularly important are the thermodynamic and kinetic data that support effective Kd models, including:

- Thermodynamic data for aqueous and mineral species
- Kinetic data for mineral species

- Ion exchange reactions and selectivity coefficients (either Gaines-Thomas or Vanselow)
- Surface complexation reactions and associated stabilities

Often, available data are not presented in terms appropriate for process-based model applications. With regard to radium, data are particularly lacking for the complexation of radium with organic matter. Speciation analyses can be run to determine how functional groups bind to radionuclides and this enables stability constants to be calculated where molecules can be broken down into small functional groups. Data are not available for radium, but it may be possible to use barium or calcium as analogues.

It is intended that AMBER will be used to couple radioactive decay chains using effective parameters.

Finally, it was noted that a PhD is currently underway, funded by Andra, on the subject of organic matter complexation.

Discussion

Work is currently focusing on the unsaturated zone and whether this zone can be represented to maintain constant conditions is currently uncertain. Ideally, the unsaturated zone should be varied over timescales linked to variations in the water table, but the availability of supporting data is considered a key limitation to this approach.

Overall, the approach to determining effective Kd was considered beneficial in that it takes account of transport across the GBIZ.

A key data requirement relates to the stability of organic matter to determine what is insoluble and what is mobile. It was noted that the Belgian HP1 approach for the near-field incorporates the unsaturated zone and may provide useful information.

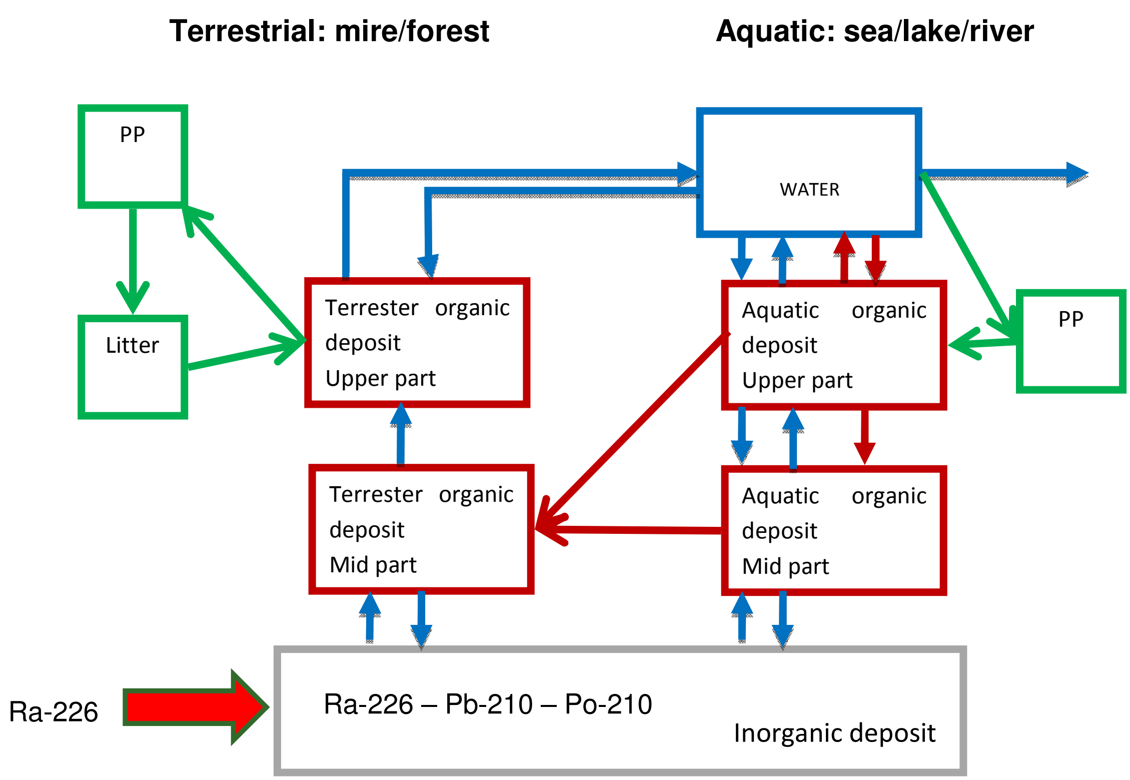
2.12 RA-226 IN A SWEDISH SAFETY ASSESSMENT (SARA NORDEN)

The Swedish safety assessment for the Forsmark site is largely based on site specific data. Calculations are made of the radioactivity concentrations in different environmental media and dose to both humans and non-human biota are estimated. The focus of the presentation was on the dose assessment methodology for humans.

A landscape model is employed that incorporates a number of interconnected landscape objects. Each object can transform during the simulation as a result of landscape evolution. Continuous landscape evolution is incorporated into the model rather than discrete events being assumed. Input to the biosphere can result from the direct use of groundwater (well abstraction) and/or direct discharge to the biosphere. The model is run on a unit release rate scenario.

Agricultural land is treated as a separate case from other landscape objects since human influence will be greater – future generations may have different lifestyles such as hunter / gatherer. Alternatively, land areas may be drained for the purposes of crop cultivation. For each alternative, a similar level of probability is assigned. However, for cultivated crops, current practices are assumed. For example, irrigation is only applied to vegetables and not cereal crops as is current practise.

The basic model is presented below.

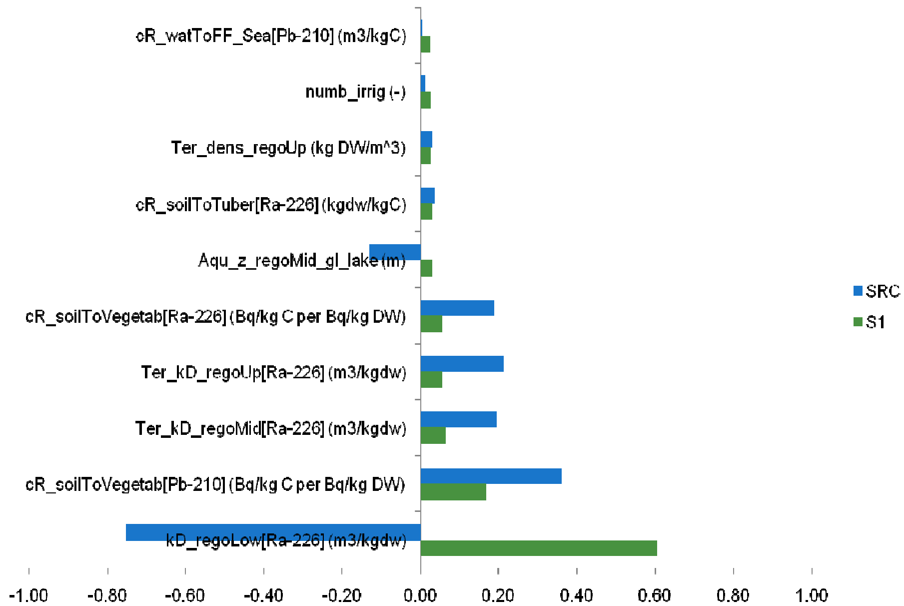


Boxes outlined in red represent organic material. Green boxes relate to primary production whereas blue boxes relate to water. Green arrows represent biotic processes. The model assumes that both Pb-210 and Po-210 are not released from the geosphere but rather appear in the biosphere as a result of in-growth from Ra-226. In order to be conservative, losses of Rn-222 are not considered. For each compartment there is a balance equation that accounts for transfers between compartments (including input, loss and any in-growth from parent radionuclides).

Equilibrium in activity concentrations of Ra-226, Po-210 and Pb-210 occurs within few thousand years.

Doses to the most exposed group vary considerably over time. The largest potential for high doses appears at the end of the simulation period, when an object is entirely terrestrial following transition from sea through lake to agricultural land. Of the three radionuclides presented, Po-210 has the lowest contribution to dose. For all three radionuclides, ingestion is the most important dose pathway with vegetables being the most important food group as a result of irrigation assumptions.

Uncertainty analysis has been performed in number ways. For deterministic cases, each input parameter has one value, based on site-specific data where available. For probabilistic scenarios, each parameter is assigned a probability distribution function (pdf). A skewed distribution of results occurs for probabilistic scenarios compared with deterministic cases as a result of skewed input parameter distributions. Those contributing most to uncertainty have been identified:



S1 indicates the percent uncertainty explained by a parameter. In this example, the majority of uncertainty is associated with the Kd for radium. No site-specific Kd values are currently available so literature data have been considered. However, a large range in Kd values is evident.

It is intended that site data will be obtained for radium, but alternative approaches have been considered such as the use of strontium as an analogue. However, use of strontium as an analogue would only occur where co-precipitation of radium with barium can be excluded. Where co-precipitation occurs, soil solution concentrations of radium will be estimated and an alternative model applied to simulate root uptake from soil solution.

The current approach for modelling plant uptake is based on concentration ratios, which relate plant concentration to the concentration in soil. For uptake to plants from activity in soil solution the form of uptake by plants is required. Where passive uptake occurs, transpiration can be used to determine uptake. However, for active transport nutrient modulated uptake is assumed. Radium is an analogue of calcium and, since calcium is taken up by passive mechanisms, a simple transpiration driven model can be used. Approaches for different radionuclides are presented in the following table.

Nuclide	Approach to root uptake	Analogue
Cs	Nutrient modulated	K
I	Nutrient modulated	I
Se	Nutrient modulated	S
Tc	Nutrient modulated	NO_3, SO_4, MoO_4
Ni	Nutrient modulated/ Driven by transpiration	Ni
Ra	Nutrient modulated/ Driven by transpiration	Ca
Sr	Nutrient modulated/ Driven by transpiration	Ca
Cl	Nutrient modulated/ Driven by transpiration	Cl
Np	Driven by transpiration	
Pu	Driven by transpiration	
Am	Driven by transpiration	
Th	Driven by transpiration	
U	Driven by transpiration	

Discussion

Concern was raised with regard to the potential application of the root uptake methodology to alternative radionuclides since, if using transpiration flux and the concentration of a radionuclide in solution, issues could arise where not all forms of a radionuclide in solution are equally bioavailable. Such an occurrence would relate to uranium. However, it was noted that the approach is only currently intended to be applied to radium as a means of addressing uncertainty associated with radium Kd values.

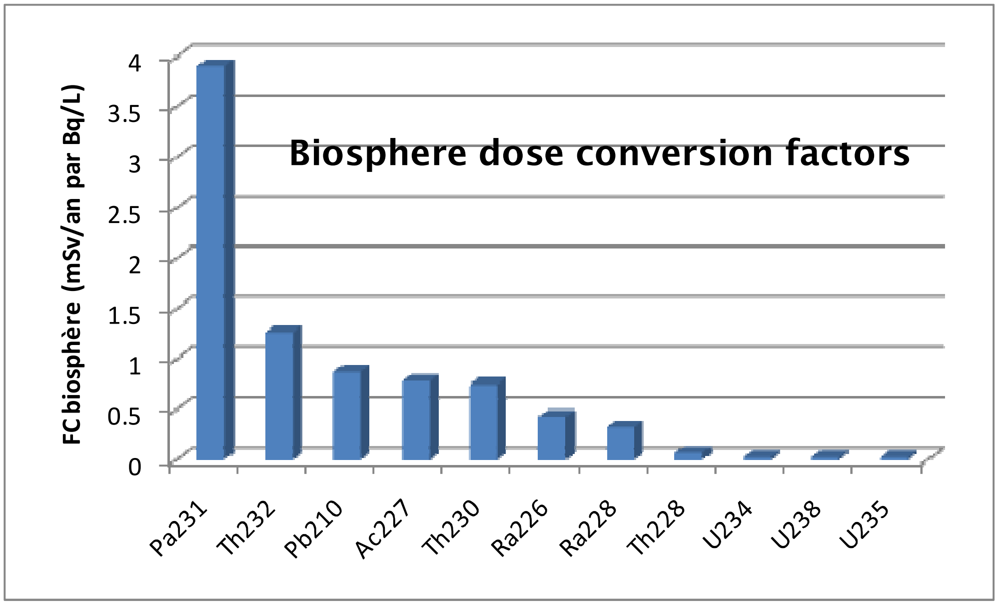
2.13 TREATMENT OF RADIUM IN THE ANDRA BIOSPHERE IMPACT ASSESSMENT (ACHIM ALBRECHT)

Key data required for human dose assessments include soil concentrations, food concentrations, information relating to human behaviour and regulatory dose limits.

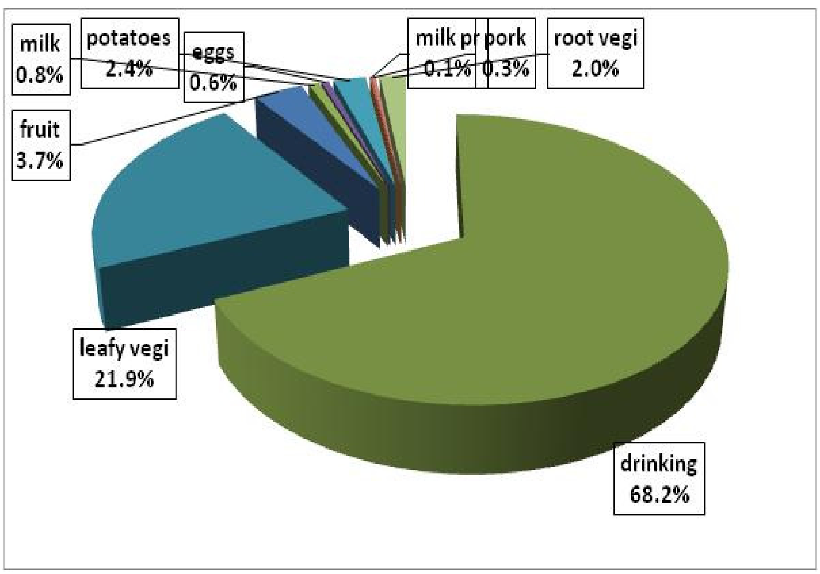
In order to calculate soil concentrations, transport of radionuclides across the GBIZ must occur. The simplest assumption in this regard is the abstraction of groundwater from a well. However, for near-surface radioactive waste repositories such a scenario is unrealistic – modelling of the upward flux of radionuclides into the biosphere is required.

On the basis of dose factors, inhalation, ingestion and external radiation must be considered for dose assessment relating to Ra-226.

Biosphere dose conversion factors (BDCFs) have been calculated for radium and other radionuclides (based on a 1 Bq/l concentration in well water) as indicated in the figure below (for a 10,000 year contamination period).



Drinking water and leafy vegetables are the most important ingestion pathways with respect to radium, with a 68% and 22% contribution to dose, respectively.



Under a stochastic analysis approach, a range of BDCFs are calculated that can span two orders of magnitude. High BDCFs are largely associated with human behaviour assumptions, which become more conservative for the future, so increasing the calculated BDCF.

Uncertainty analysis has identified Kd, precipitation, re-suspension and inhalation as important factors. Time budget can also be important (the time spent in a contaminated area).

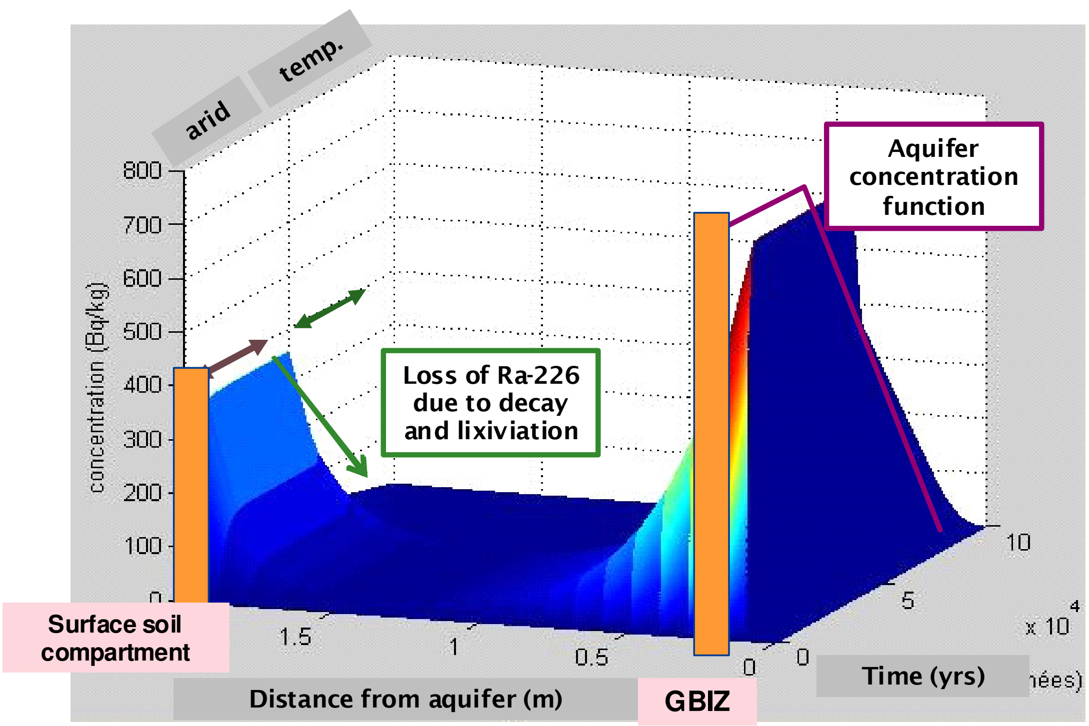
For a near-surface disposal site, irrigation water can be assumed as a source. However, if destructive effects on the disposal facility occur, upward transport across the GBIZ is more likely.

For uptake of radium into plants, the key parameters are the soil to plant transfer factor, the total soil concentration and the root depth distribution.

The concentration of radium in pore water is a function of depth and time. Advection downwards is dependant upon climate. Upward advection is more difficult to model – it is assumed that if there is not enough precipitation to support plants then plant water is obtained from below. Diffusion is integrated into the calculations, which include decay and in-growth. The distribution of roots with depth is important and the Andra model therefore divides the rooting zone into multiple compartments. Root uptake can occur throughout the entire rooted soil column. The model enables the effect of different root depth distributions on radionuclide uptake to be analysed. Recycling of plant material is also considered whereby radionuclides can be returned to surface soil following plant death.

The transfer factor is not changed as a function of depth. However Kd can be altered. For shallow-rooted plants this is not an issue as there will be a high Kd for radium. However, deep-rooted plants will be able to transport radium to the surface.

Climate change can affect the upward advective movement of radionuclides. Such transport becomes important for arid and semi-arid environments as demonstrated in the figure below. Where more rapid upward transport occurs, greater concentrations in plants are calculated.



2.14 TECHNICAL SOLUTIONS FOR THE IMPLEMENTATION OF THE FAVL DISPOSAL CONCEPT (FANNY GERARD)

Following the presentation by Achim Albrecht, Fanny Gerard gave a short presentation on the Andra FAVL programme.

The FAVL disposal project aims to apply knowledge to improve disposal safety for graphite and radium-bearing wastes. Initially the project considered the characteristics of the waste in order to design the disposal concept around this.

For graphite wastes, underground excavation and disposal at an intermediate depth with an intact cover is the preferred approach.

For radium, an open pit design with an earthwork cover is anticipated. The pit would be constructed on the basis of a sloping excavation within clay soil until the required disposal depth was achieved. Once waste was emplaced, earthworks would be undertaken to restore the pit back to natural ground level. The cover would be comprised of compacted layers that are then covered with a material that would prevent root and animal penetration. Consideration is being given as to the most appropriate cover material to employ, and also as to whether additional materials should be added as a means of preventing radium release (such as calcium).

Discussion

Radium-bearing waste is classified as low activity long-lived waste, which has a radium concentration of around 200 Bq/g. In total there are some 60,000 to 100,000 cubic metres of this waste. For some of the waste, co-precipitation of radium with barium sulphate has been performed and consideration is being given as to whether this should be recommended for the remaining waste.

Waste will largely be conditioned in metal boxes with any gaps filled with concrete to maintain mechanical strength. However, conditioning will depend to an extent on the range of radionuclides present. Where uranium is present, concrete will be used as this effectively prevents uranium migration. For other radionuclides, concrete may not be the most appropriate material however.

Conditioned wastes will be placed directly on the clay floor of the repository. No additional ground cover is planned. The maximum depth of the near surface repository will be 15 m.

It was noted that the diversity of surface cover barriers can help constrain release. The point was made that an assessment could be undertaken to determine how good containment would need to be with respect to constraining radium releases, within the context of the local hydrology, in order to meet regulatory constraints. However it was argued that in France there is a requirement to design the best possible facility and then evaluate impact rather than designing a facility to meet minimum safety standards. The principles of optimisation and justification are key.

The point was raised that, by preventing root ingress, gas release would be prevented. The cap would also potentially prevent water ingress although entry of the repository from the side could occur, which would affect hydrology. The uncoupling of the biosphere and geosphere was therefore argued against. However, the low production rate of gas in this waste stream and the limited availability of water for dissolution is considered to limit the gas issue. The use of large cobbles as a means of preventing intrusion was suggested (root access would be limited due to the low water retention in such material).

3. KEY DATA GAPS AND UNCERTAINTIES

A number of gaps and uncertainties were identified as a result of the presentations on radium environmental behaviour and modelling:

- Data relating to the accumulation of radium in soil, especially in clay and organic substrates, are limited, including thermodynamic data for interactions of radium and organic matter, which limits the ability to calculate uptake into the food chain, but also limits the ability to estimate external and inhalation doses.
- Data relating to the accumulation of radium in sediments are limited, with similar implications to the above.
- Environmental change may affect radium mobility. For example, changes in redox conditions or interactions with barium can have implications for the bio-availability of radium such that changes in the vertical pattern of soil properties could affect the distribution of radium estimated in models with multiple soil layers. For near-surface disposal facilities, the use of concrete could affect interactions between calcium and radium and such effects could warrant further consideration.
- Transfer to crops is highly variable and is dependant upon both soil and plant properties. It was suggested that soil accumulation and uptake to crops are processes that are overly-simplified. The range of soil to plant transfer factors is very large and there is a need to understand the considerations driving these differences to ensure appropriate parameter values can be selected for specific sites.

Data gaps and uncertainties associated with radium have implications for the distribution of the daughter products Pb-210 and Po-210, which have the potential to dominate radiological impacts.

4. DISCUSSION AND IDEAS FOR RESOLUTION OF KEY DATA GAPS AND UNCERTAINTIES

A number of suggestions were made as to approaches that could be taken to address the key data gaps and uncertainties identified. These are outlined below.

4.1 SOIL TO PLANT TRANSFER

There is relatively limited source data available for analysis on soil to plant transfers of radium, which become even more limited when the availability of necessary supporting soil data are considered. Work has previously been done to analyse the available data, however discrepancies have been identified in the results of these interpretations (see presentation by Hildegarde Vandenhove, Section 2.9). It was therefore recommended that work should be developed to resolve these discrepancies.

The benefit of the original data being made more widely available was also highlighted.

In the controlled experiments presented by Hildegarde Vandenhove, linear regressions between radium uptake into plants and soil / plant parameters were very strong. However, when the broader literature was consulted no overall effect of soil properties on radium uptake into plants was observed when all data were considered together. However, some specific factors such as organic matter and pH significantly explained the variation observed when specific crop types were considered. There is therefore a need to identify what is uncontrolled (or not specified) in field studies that does not occur under controlled conditions (although it was noted that the selection of soils on the basis of contrasting properties may have helped in the identification of correlations – more mixed soils would not be expected to show such strong correlations). If anomalies were observed that relate to a particular property then it may be possible to derive a targeted research programme to see whether results can be reproduced and whether robust trends can be observed.

4.2 ANALOGUE APPLICATIONS

A number of analogues have been identified for radium including barium, calcium and potentially strontium. These analogues all form one elemental series and the possibility of reviewing all elements in the series for analogous behaviour was suggested. It was considered that historical data should be available for such a task to be undertaken.

4.3 REVIEW OF CORRELATIONS BETWEEN RADIUM IN THE ENVIRONMENT AND RADIUM IN PEOPLE

An extended review and analysis could be undertaken, taking account of temporal and spatial influencing factors, to determine whether a correlation exists between radium in soils and in people. Such a review could help in the justification of parameter value selection. The possibility of conducting radon exhalation measurements under radon-free air conditions was suggested. Such measurements have been performed in relation to occupational health measurements. However, the limit of detection was not known so routine application to populations is uncertain.

An expert working group has been working to put in place a cancer register for uranium mining over the last three years. This register may be a source of useful information. Similarly, information relating to Russian mining sites and activity concentrations in people may be useful. It is thought that data exist for uranium in human tissues, but the availability of data for radium is less certain and would require investigation. Caution would be required in the application of the above data sets. Problem areas are likely to have triggered epidemiological / cancer studies thus detection rates would be greater due to the focus of the study and death rates might be lower due to subsequent treatment. Consideration would need to be given as to the detection limits for radium and whether these would be appropriate for measuring concentrations in people under normal environmental conditions.

4.4 EFFECTIVE KD

The reactive chemistry approach to dealing with Kd presented by Lara Duro received much interest. However, a limiting factor in being able to progress this further will be the availability of data to support the approach.

The presentation by Lara Duro indicated that radium is not strongly hydrolysed and so can enter the clay structure. There is a need to understand the role that organic matter plays in controlling radium behaviour before a large-scale project is undertaken. The potential benefit of Steve Sheppard and Hildegarde Vandenhove consulting on the effective Kd project was observed.

Currently the focus is on the reaction characteristics of radionuclides with active groups in fulvic and humic substances. However there is a need to consider whether other organic compounds in soils that may be present at lower concentrations will influence this behaviour since in long-term assessments, the organic matter content of soils will vary and how this influences radium behaviour may be important.

4.5 CLAY SOIL PROFILES

It was noted from the presentation by Hildegarde Vandenhove that clay soils, which are particularly important for repository sites, are among the least documented, which is a potentially important data gap. It was noted that it may be interesting to look at soil profiles above a near-surface disposal site located in clay soils. Such a study could give valuable insight into changes in soil profile relating to soil progression over time. Any gradients observed could have a large impact on radium transport.

4.6 MODELLING THE SOIL ROOT COMPARTMENT

The multi-barrier systems for near-surface disposal facilities could lead to interest in simple multi-layer models. However, the root system of plants plays an important role in the cycling of some radionuclides within surface soils, particularly radium. The root system as a biological compartment should therefore be included within models. The dose implications of including a multi-layered soil rooting zone on dose calculations are not clear and the suggestion was made that this be investigated.

4.7 BIOSPHERE DOSE CONVERSION FACTORS

Biosphere dose conversion factors (BDCFs) are often calculated in assessments and the suggestion was made that these could be reviewed in relation to identifying what drives differences between BDCFs for different sites. Not only could explanation be given as to what leads to observed differences, but important distinctions between different approaches employed could be identified.

4.8 UNDERSTANDING CONTAINMENT REQUIREMENTS

It may be interesting to ask the question ‘how slow would a release to the biosphere have to be from near-surface facilities in order to be able to safely dispose of radium-bearing waste?’ The question aims to identify how efficient waste containment needs to be (i.e. identifying required containment factors) for a range of biospheres.

Part of our role within BIOPROTA is to help in identifying the best ways for presenting information to members of the public to ensure that information is presented in an understandable context. The objective of the task outlined would be to support public information dissemination. The objective would not be to ascertain minimum containment requirements to meet regulatory criteria.

4.9 EFFECT OF CLIMATE AND ENVIRONMENTAL CHANGE ON RADIUM CONTAINMENT AND MOBILISATION

Changes in environmental conditions (temperature, precipitation, vegetation etc) have the potential to affect soil pedogenesis (e.g. at the surface of waste repositories) which influence the behaviour of radium in soils and uptake into plants. It was therefore suggested that a review of what factors would be most important in driving radium re-mobilisation in soils and clays and to what degree they impact upon radium recycling in typical ecosystems could be undertaken. Such a review may also help identify where additional research is required to address key data gaps and uncertainties.

APPENDIX A. LIST OF PARTICIPANTS

Participant	Affiliation	Presentation / Role
Abdesselam Abdelouas	Subatech, France	Radium sorption on soils – presented in his absence by Graham Smith
Achim Albrecht	Andra, France	Treatment of radium in the Andra biosphere assessment
Anne Battani	Andra, France	Participant
Benoît Madé	Andra, France	Participant
Elisabeth Leclerc	Andra, France	Participant
Eric Viollier	IPGP, France	Participant
Fanny Gerard	Andra, France	Participant
Graham Smith	GMS Abingdon Ltd, UK	Overview of data interests based on example assessments for waste management BIOPROTA Technical Secretariat
Hildegarde Vandenhove	SCK.CEN, Belgium	Predicting radium availability and plant uptake: proposal for new best estimates of Kd and TF for radium
Jean-Charles Robinet	Andra, France	Participant
Karen Smith	RPS, UK	BIOPROTA Technical Secretariat
Keiko Tagami	NIRS, Japan	Estimation of soil to plant transfer factors for radium-226
Koen Mannaerts	FANC, Belgium	Participant
Lara Duro	Amphos21, Spain	Data requirements to determine effective distribution coefficients for radium in soils
Laura Marang	EdF, France	Participant
Laurent Paurcelot	IRSN, France	Participant
Leandro Magro	ISPRA, Italy	Participant
Marie-Odile Gallerand	IRSN, France	Review of the radiological characteristics of the environment around a former uranium mine site in France: the case of the Saint Pierre site in the Cantal district of France.

Participant	Affiliation	Presentation / Role
Marion Jeambrun	IRSN, France	Uranium and its decay products in the human food chain
Michael Descostes	Areva, France	Participant
Mike Thorne	Mike Thorne & Associates, UK	The behaviour of radium and its progeny in mammals
Rachid El Mrabet	CNESTEN, Morocco	Environmental behaviour of radium-226 in aquatic and terrestrial ecosystems
Rafael Garcia-Tenorio	University of Seville, Spain	Overview of the effect of phosphogypsum amendments in agricultural soils
Sara Grolander	SKB, Sweden	Participant
Sara Norden	SKB, Sweden	Ra-226 in a Swedish safety assessment
Scott Altmann	Andra, France	Participant
Vannapha Phrommavanh	Areva, France	Participant
Victor Martz	IPGP, France	Participant
Yves Thiry	Andra, France	Radium mobility and cycling in re-vegetated mining debris

APPENDIX B. REFERENCES

The following references were provided within workshop presentations and are reproduced here for information:

- Abril J.M., García-Tenorio R., Enamorado S., Hurtado M.D., Andreu L.and Delgado A. (2008). The cumulative effect of three decades of phosphogypsum amendments in reclaimed marsh soils from SW of Spain: ^{226}Ra, ^{238}U and Cd in soils and tomato fruits. Science of the Total Environment 403: 80-88.

- Abril J.M., García-Tenorio R., Periañez R., Enamorado S,M., Andreu L. and Delgado A. (2009). Occupational dosimetric assessment (inhalation pathway) from the application of Phosphogypsum in agriculture in South West of Spain. Journal of Environmental Radioactivity 100: 29-34.

- Abril J.M., García-Tenorio R., and Manjón G. (2009). Extensive radioactive characterization of a phosphogypsum stack in SW of Spain: ^{226}Ra, ^{238}U, ^{210}Po concentrations and ^{222}Rn exhalation. Journal of Hazardous Materials 164: 790-797.

- Abril J.M., García-Tenorio R., Enamorado S., Polvillo O., Delgado A., Andreu L., Hurtado S., Villa M., Periánez R. and Manjón G. (2008). Radiological assessment of the agriculture use of phosphogypsum in SW Spain. Results of a three-year field experiment. Proceedings 5th International Symposium on Naturally Occurring Radioactive Materials (NORM V), Sevilla, Marzo 2007. IAEA Proceedings Series pages 297-306. ISBN 978–92–0–101508–2.

- Albrecht, A., 2008. Déchets radifères : Impact des différentes voies d'exposition sur le facteur de conversion biosphère des radionucléides des chaînes de désintégration d'U-238, U-235 et Th-232. Report Andra N° F.NT.ASTR.08.0043, Châtenay-Malabry.

- Albrecht, A. and Miquel, S., 2009. Extension of sensitivity and uncertainty analysis for long term dose assessment of high level nuclear waste disposal sites to uncertainties in the human behaviour. J. Environ. Radioactivity, 101: 55-67.

- Alexander F W, Clayton B E and Delves H T (1974). Mineral and Trace Metal Balances in Children receiving Normal and Synthetic Drugs. Quarterly Journal of Medicine, 43, 89-111.

- Amaral E C S, Carvalho Z L and Godoy J M (1988). Transfer of ^{226}Ra and ^{210}Pb to Forage and Milk in a Brazilian High Natural Radioactivity Region. Radiat. Prot. Dosim., 24(1/4), 119-121.

- Anthony D S, Davis R K, Cowden R N and Jolley W P (1956). Experimental Data Useful in Establishing Maximum Single and Multiple Exposure to Polonium. In: Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, 13, 215-218, United Nations, New York.

- ATSDR (1999). U.S. Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Lead, July 1999.

- Brown J and Simmonds J R (1995). FARMLAND: A Dynamic Model for the Transfer of Radionuclides through Terrestrial Foodchains. National Radiological Protection Board Report NRPB R273, Chilton, UK.

- Bruenger F W, Smith J M, Atherton D R, Jee W S S, Lloyd R D and Stevens W (1983). Skeletal Retention and Distribution of Ra-226 and Pu-239 in Beagles Injected at Ages Ranging from 2 days to 5 years. Health Phys., 44 (Suppl. 1), 513-527.
- Bulman R A, Ewers L W and Matsumoto K (1995). Investigations of the Potential Bioavailability of ^{210}Po in Some Foodstuffs. Sci. Total Environ., 173-174, 151-8.
- Bunzl K and Kracke W (1984). Distribution of Pb-210, Po-210, Stable Lead and Fallout Cs-137 in Soil, Plants and Moorland Sheep of a Heath. Science of the Total Environment, 39, 143-159.
- Burns B, Clulow F V, Cloutier N R, Davé N K and Lim T P (1987). Transfer Coefficient of Ra-226 from Food to Young Weaned Meadow Voles, *Microtus Pennsylvanicus*, in the Laboratory. Health Phys., 52, 207-211.
- Campbell J E, Robajdek E S and Anthony D S (1956). The Metabolism of Ac-227 and its Daughters Th-227 and Ra-223 by Rats. Radiat. Res., 4, 294-302.
- Chen X (1987). Body Content of some Naturally Occurring Radioisotopes in Chinese People. NRPB Bulletin, 84, 13-20.
- Church B W (2001). Review of Relevant Grazing Studies in Arid Areas addressing Am, Pu, U & Ra, Cs, Sr and Co Radioisotopes. BWC Enterprises, Inc., Logandale, NV 89021, August-September, 2001.
- Conrad M E and Barton J C (1978). Factors affecting the Absorption and Excretion of Lead in the Rat. Gastroenterology, 74, pp. 731-740.
- Della Rosa R J, Thomas R G and Stannard J N (1955). University of Rochester Report UR-392.
- Ewers L W, Ham G J and Wilkins B T (2003). Review of the Transfer of Naturally Occurring Radionuclides to Terrestrial Plants and Domestic Animals. National Radiological Protection Board Report NRPB-W49, Chilton, U.K.
- Fell T P, Harrison J D and Leggett R W (2001). A Model for the Transfer of Alkaline Earth Elements to the Fetus. Radiat. Prot. Dosim., 95(4), 309-321.
- Fink R M (1950). Biological Studies with Polonium, Radium and Plutonium. McGraw-Hill, London.
- Gerber G B and DeRoo J (1974). Absorption of Radioactive Lead (Pb-210) by Different Parts of the Intestine in Young and Adult Rats, Environmental Physiology and Biochemistry, 5, 314-318.
- Gerber G B, Léonard A and Jacquet P (1980). Toxicity, Mutagenicity and Teratogenicity of Lead. Mutation Research/DNA Repair, 76, 115-141.
- Gvozdanovic S (1985). Empirical Relationship for Skeletal Utilisation and Discrimination of Ca, Sr, Ba and Ra in Adult Man, Pig, Bovine and Dog. Nutr. Res. (Suppl. 1), 281-284.
- Haas G, Schupfner R and Müller A (1995). Transfer of Natural and Man Made Radionuclides from Plants to Roe Deer and Farm Animals. J. Radioanal. Nucl. Chem., 194(2), 269-276.

- Harrison J D, Leggett R W, Noβke D, Paquet F, Phipps A W, Taylor D M and MétivierH (2001). Reliability of the ICRP's Dose Coefficients for Members of the Public, II: Uncertainties in the Absorption of Ingested Radionuclides and the Effect on Dose Estimates. Radiation Protection Dosimetry, 95, 295-308.

- Harrison J D, Smith T J and Phipps A W (2003). Infant Doses from the Transfer of Radionuclides in Mothers' Milk. Radiat. Prot. Dosim., 105(1-4), 251-256.

- Hill C R (1965). ^{210}Polonium in Man. Nature, 208, 423-428.

- Hogan K, Marcus A, Smith R, White P. (1998). Integrated Exposure Uptake Biokinetic Model for Lead in Children: Empirical Comparisons with Epidemiologic Data. Environmental Health Perspectives, 106 (Supplement 6), pp. 1557-1567, December 1998.

- Holtzman R B, Urnezis P W, Padova A and Bobula C M III (1979). Contamination of the Human Food Chain by Uranium Mill Tailings Piles. Argonne National Laboratory, Illinois, USA, NUREG/CR-0758, ANL/ES—69.

- Hunt G J and Allington D J (1993). Absorption of Environmental Polonium-210 by the Human Gut. J. Radiol. Prot., 13 (2), 119-126.

- IAEA (1994). Handbook of Parameter Values for Prediction of Radionuclide Transfer in Temperate Environments. Technical Report Series No 364, International Atomic Energy Agency, Vienna, 1994.

- IAEA (2010). Handbook of Parameter Values for Prediction of Radionuclide Transfer in Temperate Environments. Technical Report Series No 472, International Atomic Energy Agency, Vienna, 2010.

- ICRP (1973). Alkaline Earth Metabolism in Adult Man. ICRP Publication 20, Pergamon Press, Oxford.

- ICRP (1979-81). Limits for Intakes of Radionuclides by Workers. ICRP Publication 30, Parts 1-3. Annals of the ICRP, 2(3/4), 4(3/4), 6(2/3).

- ICRP (1993). Age-dependent Doses to Members of the Public from Intakes of Radionuclides: Part 2 – Ingestion Dose Coefficients, ICRP Publication 67, Annals of the ICRP, 23(3/4).

- ICRP (1994). Human Respiratory Tract Model for Radiological Protection, ICRP Publication 66, Annals of the ICRP, 24(1-3).

- Kauranen P and Miettinen J K (1967). ^{210}Po and ^{210}Pb in Environmental Samples in Finland. In: Radioecological Concentration Processes (Eds. Aberg B and Hungate F P), 275-280, Proc. Symp. Stockholm, April 25-29, 1966, Pergamon Press, Oxford.

- Kendall G M, Harrison J D and Fell T P (1988). Report of the Nuclear Energy Agency Expert Group on Gut Transfer Factors: Implications for Dose per Unit Intake. Radiation Protection Dosimetry, 25(1), 59-65, 1988.

- Khademi B and Mahdavi A (1974). Increase of Ra-226 in the Water Intake by Sheep. Health Phys., 27, 522-524.

- Kirchmann R, Lafontaine A, Van den Hoek J and Koch G (1972). Comparison of the Rate of Transfer to Cow Milk of Ra-226 from Drinking Water and Ra-226 Incorporated into Hay. Biologie Comptes Rendus, 166, 1557-1562.
- Landinskaya L A, Parfenov Y D, Popov D K and Federova A V (1973). ^{210}Pb and ^{210}Po Content of Air, Water, Foodstuffs and the Human Body. Arch. Environ. Health, 27, 254-258.
- Lapham S C, Millard J B and Samet J M (1989). Health Implications of Radionuclide Levels in Cattle Raised near U Mining and Milling Facilities in Ambrosia Lake, New Mexico. Health Phys., 56, 327-340.
- Leggett R W (1992). A Generic Age-Specific Biokinetic Model for Calcium-like Elements. Paper presented at the Workshop on ‘Age-dependent Factors in the Biokinetics and Dosimetry of Radionuclides’, Schloß Elmau, Germany 5-8 November 1991, Radiation Protection Dosimetry, 41, 183-198.
- Leggett R (1993). An Age-Specific Kinetic Model of Lead Metabolism in Humans. Environmental Health Perspectives, 101(7), 598-616, December 1993.
- Leggett R W and Eckerman K F (2001). A Systemic Biokinetic Model for Polonium. Sci Total Environ., 275(1-3), 109-25.
- Linsalata P, Morse R, Ford H, Eisenbud M, Penna Franca E, de Castro M B, Lobao N, Sachett I and Carlos M (1989). Transport Pathways of Th, U, Ra and La from Soil to Cattle Tissues. J. Environ. Radioactivity, 10, 115-140.
- Lloyd R D, Mays C W, Atherton D R, Taylor G N and van Dilla M A (1976). Retention and Skeletal Dosimetry of Injected Ra-226, Ra-228 and Sr-90 in Beagles. Radiat. Res., 66, 274-287.
- Lloyd R D, Mays C W, Taylor G N, Atherton D R, Bruenger F W and Jones C W (1982). Ra-224 Retention, Distribution and Dosimetry in Beagles, Radiat. Res., 92, 280-295.
- Lucas H F and Di Ferrante E R (1966). The Ra-226, Po-210 and Th-228 in Bovine Bone and Teeth. Radiat. Res., 8, 480-489.
- Mahon D C (1982). Uptake and Translocation of Naturally Occurring Radionuclides of the Uranium Series. Bull. Environ. Contam. Toxicol., 29, 697-703.
- Marcus A H (1979). The Body Burden of Lead: Comparison of Mathematical Models for Accumulation. Environmental Research, 19, 79-90.
- Mason H J (2000). A Biokinetic Model for Lead Metabolism with a view to its Extension to Pregnancy and Lactation; (1). Further validation of the Original Model for Non-Pregnant Adults. Science of the Total Environment, 246(1), pp. 69-78, 31st January 2000.
- Mays C W, Cochran T H and Jee W S (1963). Radium and Radon Retention in Rats. Health Phys., 9, 615-619.
- McDowell-Boyer L M, Watson A P and Travis C C (1980). Review of Parameters Describing Terrestrial Food Chain Transport of Lead-210 and Radium-226. Nucl. Safety, 21, 486-494.

- McInroy J F, Watters R L and Johnson J E (1972). Polonium-210 Absorption in Rats: Effects of Biological Modification. Nature New Biol., 236, 118-120.
- Miller C E and Finkel A J (1965). Radium Retention in Mice after Single Intravenous Injection. Radiat. Res., 26, 269-286.
- Milosevic Z, Horsic E, Klajic R and Bauman A (1980). Distribution of Uranium, Ra-226, Pb-210 and Po-210 in the Ecological Cycle in Mountain Regions of Central Yugoslavia. In: Radiation Protection, a Systematic Approach to Safety, Proc. 5th Int. Conf. of IRPA, Jerusalem, Israel, Vol. 2, pp.1123-1126, INLS-MF-5876, Pergamon Press, Oxford.
- Miquel, S., 2008. Développement et intégration d'un modèle de transfert sol multicouche dans le logiciel Matlab. Report Société de Calcul Mathématique, S.A. N° C.RP.FSTR.08.0009.A, Paris.
- Miquel, S., 2010. Adaptation du modèle sol multicouche pour les applications FAVL-Radifère du Programme Scientifique. Report Société de Calcul Mathématique, S.A. N° F.RP.FSTR.10.0006.A, Paris.
- Morgan J E and Beetham C J (1990). Review of Literature for Radium, Protactinium, Tin and Carbon, Nirex Safety Studies Report No. NSS/R220, UK Nirex Limited, Curie Avenue, Harwell, Didcot, Oxfordshire, OX11 0RH.
- Morrow P, Beiter H, Amato F and Gibb F R (1980). Pulmonary Retention of Lead: An Experimental Study in Man. Environmental Research, 21, 373-384.
- Muller W A (1971). Studies on Short-lived Internal Alpha Emitters in Mice and Rats, Part 1. Ra-224, Int. J. Radiat. Biol., 20, 27-38.
- Mykkänen H and Wasserman R H (1981). Gastrointestinal Absorption of Lead (^{203}Pb) in Chicks: Influence of Lead, Calcium and Age. Journal of Nutrition, 111, 1757-1765.
- Naylor G P L, Bonas H E, Haines J W, Ham G J, Harrison J D, Sundaram S and Dayan A D (1991). The Gastrointestinal Absorption and Tissue Distribution of Alpha-emitting Actinide Isotopes and Polonium-210. The British Nuclear Energy Society Conference: Occupational Radiation Protection, Guernsey, Thomas Telford, London, pp.291-296.
- Naylor G P L, Haines J W, Whysall K, Bonas H E and Harrison J D (1993). Measurements of the Gastrointestinal Absorption and Tissue Distribution of Plutonium, Americium and Polonium in Experimental Animals. The Science of the Total Environment, 130/131, 429-435.
- NEA/OECD (1988). Committee on Radiation Protection and Public Health, Report of an Expert Group on Gut Transfer Factors. NEA/OECD, Paris.
- Nelson N S and Rust J H (1967). Distribution of and Discrimination against Naturally Occurring Ra-226 in Swine. In: Aberg, B and Hungate, F P (Eds), Radioecological Concentration Processes, Proc. Int. Symp. Stockholm, April 1966, pp.573-588, Pergamon Press, Oxford.
- Ng Y C, Colsher C S, Quinn D L and Thompson S E (1977). Transfer Coefficients for the Prediction of the Dose to Man via the Forage-Cow-Milk Pathway from Radionuclides Released to the Biosphere, UCRL-51939, Lawrence Livermore Laboratory, University of California, Livermore, California 94550.

- Ng Y C, Colsher C S and Thompson S E (1979). Transfer Factors for Assessing the Dose from Radionuclides in Agricultural Products, IAEA-SM-237-54.
- Ng Y C, Colsher C S and Thompson S E (1982). Transfer Coefficients for Assessing the Dose from Radionuclides in Meat and Eggs, NUREG/CR-2976.
- Niemi A, Venalainen E R, Hirvi T, Hirn J and Karppanen E (1991). The Lead, Cadmium and Mercury Concentrations in Muscle, Liver and Kidney from Finnish Pigs and Cattle during 1987-1988. Zeitschrift für Lebensmitteluntersuchung und -forschung A, 192(5), 427-9, May 1991.
- O'Flaherty E J (1998). A Physiologically Based Kinetic Model for Lead in Children and Adults. Environmental Health Perspectives, 106 (Supplement 6), 1495-1503, December 1998.
- Palminger Hallén J, Norrgren L and Oskarsson A (1996). Distribution of Lead in Lactating Mice and Suckling Offspring with Special Emphasis on the Mammary Gland. Archives of Toxicology, 70(3-4), 237-243, January 1996.
- Parks N J, Pool R R, Williams R J and Wolf H G (1978). Age and Dosage-level Dependence of Ra Retention in Beagles. Radiat. Res., 75, 617-632.
- Pokarzhevskii A D and Krivolutskii D A (1997). Background Concentrations of ^{226}Ra in Terrestrial Animals. Biogeochem., 39, 1-13.
- Polig E, Lloyd R D, Bruenger F W and Miller S C (2004). Biokinetic Model of Radium in Humans and Beagles. Health Phys., 86(1), 42-55.
- Quarterman J and Morrison E (1978). The Effect of Age on the Absorption and Excretion of Lead. Environmental Research, 17, pp. 78-83.
- Sansom B F and Garner R J (1966). The Metabolism of Radium in Dairy Cows. Biochem. J., 99, 677-681.
- Smith A D and Thorne, M C (1986). Pharmacodynamic Models of Selected Toxic Chemicals in Man: Volume 2 – Routes of Intake and Implementation of Pharmacodynamic Models, MTP Press, Lancaster.
- Smith F A, Morrow P E, Gibb F R, Rosa R J D, Casarett L J, Scott J K and Stannard J N (1961). Distribution and Excretion Studies in Dogs Exposed to an Aerosol containing Polonium-210. Am. Ind. Hyg. Ass. J., 22, 201-208.
- Smith T J, Phipps A W, Fell T P and Harrison J D (2003). Transfer of Alkaline Earth Elements in Mothers' Milk and Doses from ^{45}Ca, ^{90}Sr and ^{226}Ra. Radiat. Prot. Dosim., 105(1-4), 273-277.
- Stannard J N and Cassarett G W (1964). Metabolism and Biological Effects of an Alpha-emitter, Polonium-210. Academic Press, New York.
- Staples C R, Umana R, Strickler J A, Shibles D, Hayen M J, Hissem C D, Lough D S and Demorest D L (1994). Radionuclides in Milk of Dairy Heifers Raised on Forages Harvested from Phosphatic Clay Soils on Reclaimed Mined Land. J. Environ. Qual., 23, 663-667.

- Staven L H, Rhoads K, Napier B A and Strenge D L (2003). A Compendium of Transfer Factors for Agricultural and Animal Products. Pacific Northwest National Laboratory Report PNNL-13421.

- Szerbin P and Popov D K (1988). Transfer of Radium-228, Thorium-228 and Radium-226 from Forage to Cattle and Reindeer. J. Environ. Radioactivity, 8, 129-141.

- Taylor D M, Bligh P H and Duggan M H (1962). The Absorption of Calcium, Strontium, Barium and Radium from the Gastrointestinal Tract of the Rat. Biochem. J., 83, 25-29.

- Thiry, Y. and Van Hees M. (2008). Long term evolution of pH, organic matter and ^{226}radium/calcium partitioning in U-mining debris following re-vegetation with pine trees. Science of The Total Environment, 393(1): 111-117.

- Thiry Y. and Van Hees M., 2007. Forest (re)vegetation impact on radium-226 cycling in U-mining area. In Zhu, Y., Lepp, N. and Naidu, R. (Eds), Biogeochemistry of Trace Elements: Environmental Protection, Remediation and Human Health, ICOBTE 2007, Beijing, China, 15-19 July 2007, Beijing, China. p. 449-450.

- Thomas P A (1994). Dosimetry of Po-210 in Humans and Wolves in Northern Canada. Health Physics, 66, pp. 678-690.

- Thomas P A (2000). Radionuclides in the Terrestrial Ecosystem near a Canadian Uranium Mill – Part II: Small Mammal Food Chains and Bioavailability. Health Phys., 78(6), 625-632.

- Thomas P A and Gates T E (1999). Radionuclides in the Lichen-Caribou-Human Food Chain near Uranium Mining Operations in Northern Saskatchewan, Canada. Environ. Health Perspect., 107(7), 527-537.

- Thomas P, Irvine J, Lyster J and Beaulieu R (2005). Radionuclides and Trace Metals in Canadian Moose near Uranium Mines: Comparison of Radiation Doses and Food Chain Transfer with Cattle and Caribou. Health Phys., 88(5), 423-38.

- Thorne M C (2001). Modeling the Transfer of ^{226}Ra, ^{210}Pb and ^{210}Po to Farm Animals, Mike Thorne and Associates Limited Report for RWE NUKEM MTA/P0021/2001-1: Issue 1, November 2001.

- Thorne M C, Jackson D and Smith A D (1986). Pharmacodynamic Models of Selected Toxic Chemicals in Man: Review of Metabolic Data, Volume 1. MTP Press, Lancaster, 1986.

- Trampel D W, Imerman P M, Carson T L, Kinker J A and Ensley S M (2003). Lead Contamination of Chicken Eggs and Tissues from a Small Farm Flock. Journal of Veterinary Diagnostic Investigation, 15(5), 418-22, September 2003.

- Underwood E J (1977). Trace Elements in Human and Animal Nutrition, 4th Edition, Academic Press, 1977.

- Underwood, E J and Suttle, N F (1999). The Mineral Nutrition of Livestock, 3rd Edition, CABI Publishing, Wallingford, 1999 (Reprinted with corrections 2001).

- UNSCEAR (2006). Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly. United Nations, New York.

- Van Dilla M A, Stover B J, Floyd R L, Atherton D R and Taysum D H (1958). Radium (Ra-226) and Radon (Em-222) Metabolism in Dogs. Radiat. Res., 8, 417-437.
- Vandenhove, H., C. Gil-García, A. Rigol and M. Vidal. 2009. Proposal for new best estimates for the solid-liquid distribution coefficients in soils of the natural radionuclides U, Th, Ra, Pb and Po. Journal of Environmental Radioactivity, 100, 697-703.
- Vandenhove, H., G. Olyslaegers, N. Sanzharova, O. Shubina, E. Reed, Z. Shang, and H. Velasco. 2009. Proposal for new best estimates of the soil-to-plant transfer factor of U, Th, Ra, Pb and Po. Journal of Environmental Radioactivity, 100, 721-732.
- Vos G, Hovens J P and Delft W V (1987). Arsenic, Cadmium, Lead and Mercury in Meat, Livers and Kidneys of Cattle slaughtered in the Netherlands during 1980-1985. Food Additives and Contaminants, 4(1), 73-88, January – March 1987.
- Watson A P, Etnier E L and McDowell-Boyer L M (1983). Radium-226 in Drinking Water and Terrestrial Food Chains: a Review of Parameters and an Estimate of Potential Exposure and Dose. Oak Ridge National Laboratory, Oak Ridge, Tennessee, ORNL/TM-8597.
- Watters R L and McInroy J F (1969). The Transfer of Po-210 from Cattle Feed to Milk. Health Physics, 16, pp. 221-225.
- White P D, Van Leeuwen P, Davis B D, Maddaloni M, Hogan K A, Marcus A H and Elias R W (1998). The Conceptual Structure of the Integrated Exposure Uptake Biokinetic Model for Lead in Children. Environmental Health Perspectives, 106 (Supplement 6), 1513-1530, December 1998.
- Wrenn M E, Durbin P W, Howard B, Lipsztein J, Rundo J, Still E T and Willis D T (1985). Metabolism of Ingested U and Ra. Health Phys., 48, 601-633.
- Zaragoza L and Hogan K. (1998). The Integrated Exposure Uptake Biokinetic Model for Lead in Children: Independent Validation and Verification. Environmental Health Perspectives, 106 (Supplement 6), pp. 1551-6, December 1998.
- Ziegler E E, Edwards B B, Jensen R L Mahaffrey K R and Fomon S L (1978). Absorption and Retention of Lead by Infants. Pediatric Research, 12, 29-34.