

BIOPROTA

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

Improving Confidence in Long-term Dose Assessments for U-238 Series Radionuclides

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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.bioprot.org.

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.

In 2010, a U-238 series radionuclide project was established to address the U-Th-Ra part of the decay chain, with special focus on the soil-plant aspect of radionuclide behaviour in the biosphere, taking into account the effects of dynamic decay and in-growth in the decay chain.

The work programme has been supported financially and technically by ANDRA, NDA RWMD, NRPA, NUMO and NWMO. Additional technical input has been provided by CIEMAT, IRSN, the University of Bremen, and the University of Lausanne (CHUV), and the technical support team, made up from GMS Abingdon, Quintessa, Eden Nuclear and Environment, Mike Thorne and Associates, and Limer Scientific Consulting,

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EXECUTIVE SUMMARY

Many organisations have an interest in the long-term radiological impacts of U-238 series radionuclides, which may be released from radioactive waste disposal facilities. The half-life of U-238 is much longer than the stability of near-surface environments and the likely period of effectiveness of engineered and natural barriers in shallow and deep disposal facilities. Particular interest arises because of the very wide range of half-lives of the radionuclides in the series, and the degree of disequilibrium which may arise over different temporal and spatial scales.

In 2010, a U-238 series radionuclide project was established to address the U-Th-Ra members of the decay chain, with special focus on the soil-plant aspect of radionuclide behaviour in the biosphere. This report describes the activities carried out within the project including:

- The development of a generic interaction matrix that details linkages between key features and processes affecting the migration of the radionuclides of interest, U-238, U-234, Th-230 and Ra-226, in surface soils and in crops;
- The description of a range of assessment models;
- The development and modelling of two scenarios, and quantitative estimates of the U-238, U-234, Th-230 and Ra-226 concentrations in specific components of the dose assessment models (soil and plants), for agreed geosphere to biosphere release scenarios of contaminated groundwater, for a hypothetical and an actual site.
- The presentation of results to allow similarities and differences in the different modelling approaches used to evaluate the scenarios.

With respect to the key features and processes affecting the radionuclides considered in this study, they are applicable to a large number of other radionuclides. Those FEPs which were discarded from the IUR FEP matrix in Section 3 generally related to the behaviour of gas since the current study does not consider Ra-226 progeny such as Rn-222 gas.

Two types of model have been used in this study:

- conventional radionuclide transfer models with one or two soil compartments, which consider simplistic dynamics of the radionuclides within a soil profile; and
- more complex models that have the potential to account for changes in redox conditions throughout a soil profile (discretised into more than 2 layers), and the effect of redox conditions upon radionuclide behaviour (e.g. sorption).

The models are summarised in Section 4, with the technical details given in Appendix B.

The scenarios, hypothetical and site-specific, were developed specifically for this study. In the case of the hypothetical scenario, use was made of previous model intercomparison studies in the scenario definition. The set of calculations and results for different precipitation conditions, made using one of the assessment models (SAMM), illustrate the potential significance of including subsoil layers explicitly within the model, particularly as this affects the representation of upward as well as downward radionuclide movement.

For the site-specific scenario, data was collated from a range of reports and scientific publications produced over the last decade on the Los Ratones site in Spain.

Results for the site-specific scenario suggest that the calculated potential radionuclide concentrations in soil and plants vary between models by typically one order of magnitude, and up to a factor of 75. The results also suggest that U-238, U-234, Th-230 and Ra-226 show differences from secular equilibrium of the same order. The models have been developed based on data derived from a range of sites, not just for the Los Ratones site, and so it is not surprising that the range in results is comparable with that observed generally in the environment. It is clear that further characterisation data at depth, and information about the history of activities at the site, would be additionally useful in possible future work.

The more complex models are more data intensive. Further questions which could be considered include: Does that complexity make the results more useful? Does it provide greater insight? When is it necessary to use the more complex approaches? How might site-specific data be used to inform assessment models? For example, future work could consider the implications of being able to estimate or determine site-specific sorption coefficients, based upon information about the soil properties at the site.

Consideration could also be given to the potential importance, or relevance, of accounting for the natural background content of uranium series radionuclides in the soil and water upon the calculated radionuclide concentrations of soil and plants following some form of release as a result of human activities (e.g. mining activities, waste disposal).

There are other avenues of research which the future work of the U-238 series radionuclide working group could consider, dependent upon the wider interests of the group. Consideration could be given to other aspects of the terrestrial ecosystem, such as exposure pathways other than the soil-plant pathway (e.g. inhalation), impacts to non-human biota and doses to humans, or the soil-plant uptake in a non-agricultural ecosystem. If the analysis were extended to comprise freshwater systems, then consideration would need to be given to the process of the accumulation of radionuclides in the freshwater sediments (in rivers and lakes), and the subsequent transfer pathways to aquatic organisms.

This study has focussed upon the upper members of the decay chain. Future work may remain focussed upon the soil-plant system, but considering Ra-226 decay products, notably Rn-222, Pb-210 and Po-210.

CONTENTS

| | |
|---|-----------|
| PREFACE | I |
| EXECUTIVE SUMMARY | II |
| 1. INTRODUCTION | 1 |
| 1.1 Background and objective | 1 |
| 1.2 Report Structure | 2 |
| 2. URANIUM SERIES RADIONUCLIDES IN THE BIOSPHERE | 3 |
| 2.1 Behaviour of uranium series radionuclides in soil | 3 |
| 2.2 Uptake by plants | 4 |
| 3. ANALYSIS OF KEY PROCESSES FOR U-238 SERIES RADIONUCLIDES IN A SOIL-PLANT SYSTEM | 6 |
| 3.1 Interaction matrix for U-238, U-234, Th-230 and Ra-226 | 7 |
| 3.2 Process descriptions | 10 |
| 4. MODELS FOR CONSIDERATION IN THIS STUDY | 13 |
| 4.1 The CIEMAT soil redox model | 13 |
| 4.2 Andra's SAMM model | 14 |
| 4.3 The NDA RWMD Biosphere Assessment Tool | 15 |
| 4.4 SYVAC3-CC4 | 15 |
| 4.5 RESRAD-OFFSITE | 15 |
| 4.6 "Calculation Bases Mining" model (CBM) | 16 |
| 4.7 Summary of processes considered in these models | 16 |
| 5. HYPOTHETICAL SCENARIO DESCRIPTION AND DATA | 18 |
| 5.1 Scenario description | 18 |
| 5.2 Parameter values used in the SAMM model | 19 |
| 6. HYPOTHETICAL SCENARIO RESULTS | 22 |
| 6.1 Case 1: High Precipitation (723 mm y^{-1}) | 22 |
| 6.2 Case 2: Low Precipitation (431 mm y^{-1}) | 26 |
| 7. LOS RATONES SCENARIO DESCRIPTION AND DATA | 29 |
| 7.1 scenario outline | 29 |
| 7.2 LOS RATONES description | 29 |
| 7.3 Parameter Values | 37 |
| 8. LOS RATONES SCENARIO RESULTS | 43 |
| 8.1 SAMM model | 43 |

| | |
|---|-----------|
| 8.2 NDA RWMD Biosphere Tool | 48 |
| 8.3 SYVAC3-CC4 | 49 |
| 8.4 Discussion of Los ratones results | 50 |
| 9. SUMMARY AND CONCLUSIONS | 52 |
| 10. REFERENCES | 54 |
| APPENDIX A: THE IUR GENERAL INTERACTION MATRIX FOR A TERRESTRIAL ECOSYSTEM | 59 |
| APPENDIX B: TECHNICAL DETAILS OF THE MODELS CONSIDERED IN THIS STUDY | 61 |
| B.1 CIEMAT redox model | 61 |
| B.2 Andra's SAMM model | 62 |
| B.3 NDA RWMD Biosphere Assessment Tool | 65 |
| B.4 SYVAC3-CC4 | 70 |
| B.5 RESRAD-OFFSITE | 74 |
| B.6 "Calculation Bases Mining" model (CBM) | 76 |

1. INTRODUCTION

1.1 BACKGROUND AND OBJECTIVE

Many organisations have an interest in the long-term radiological impacts of U-238 series radionuclides. Particular interest arises because of the very long half-lives of some of the radionuclides in the series. These are much longer than the stability of near-surface environments and the likely period of effectiveness of engineered barriers in shallow disposal facilities. The half lives are also significant in relation to the containment that can be provided by deep geological disposal systems. Special issues arise concerning assumptions for: uranium mobility (depending on local biogeochemistry); the in-growth and decay of radioactive progeny and assumptions as to their disequilibrium in different media, below and above ground; the treatment of Rn-222 release and related exposure pathways (including indoor exposure pathways in dwellings); and the behaviour of Pb-210 and Po-210 in the surface environment.

These issues were discussed at the annual BIOPROTA meeting held in May 2009, at CIEMAT, Madrid (BIOPROTA, 2009). It was concluded there that the interest in the modelling of U-238 series radionuclides is not focused solely on deep geological disposal and that the following issues are also of relevance:

- delayed and long-term release of U-238 and daughters in groundwater resulting from shallow or near-surface disposal of uranium and radium bearing wastes;
- releases from uranium mining and mill tailing waste facilities (including water treatment plants) and other waste disposal facilities from NORM (Naturally Occurring Radioactive Material) industries; and
- residual activity at legacy sites involved in processing large volumes of NORM material.

The issues associated with modelling the behaviour of U-238 and its progeny once released into the near-surface geosphere and biosphere are generally independent of the type of facility from where they arise. There may be exceptions in relation to site-specific issues, such as release of oxyanions (nitrates) or organic matter that can influence U mobility via redox changes or organic complexation, or local downward movement of oxygen-bearing waters and differences in the source term with regard to chemical form. There is commonly a need to consider the full decay chain, or at least several members of it, in order to appropriately address disequilibrium and migration issues, including radon emanation in different environmental situations and taking account of the impacts of environmental change.

In 2010, a U-238 series radionuclide project was established focussing on the U-Th-Ra members of the decay chain. The objective was to provide a mechanism for sharing information on assessment methods and data interpretation and hence improving confidence in long-term dose assessment for U-Th-Ra releases.

This report describes activities of the project, including:

- The development of a generic interaction matrix that details linkages between key features and processes affecting the migration of the radionuclides of interest;
- The description of the computer models used in the study;
- The development and modelling of two scenarios; and

- The analysis of results to evaluate similarities and differences in the different modelling approaches used to evaluate the scenarios.

Within this study, two scenarios have been considered: one hypothetical scenario and one based on site-specific data. This study focuses on the soil-plant aspect of radionuclide behaviour in a terrestrial ecosystem; other aspects which contribute to human exposure can be considered in future studies. Thus the assessment question to be addressed by the modelling is:

What are the concentrations of U-238, U-234, Th-230 and Ra-226 in the edible parts of the harvested crops consumed by humans and animals, taking into account decay chain dynamics?

The hypothetical scenario is similar to the scenarios considered in the BIOPROTA studies on Cl-36 and C-14 (Bytwerk et al., 2011; Limer et al., 2008, 2011). The site-specific scenario is based upon information from the former uranium mine site of Los Ratones in Spain.

1.2 REPORT STRUCTURE

As a background, Section 2 provides some information on the understanding of the behaviour of uranium series radionuclides in the terrestrial biosphere. Section 3 outlines an initial FEP and interaction matrix analysis which has been carried out for the radionuclides of interest, i.e. the higher members of the decay chain up to and including Ra-226. The models used in this project are summarised in Section 4. The hypothetical scenario and the data used in the calculations are described in Section 5, and the results in Section 6. The site-specific scenario is described in Section 7, with the results in Section 8. The conclusions and summary are given in Section 9.

References are provided in Section 10. The original generic terrestrial interaction matrix from the International Union of Radioecology (IUR) is presented in Appendix A. The technical details of the models summarised in Section 4 are given in Appendix B.

2. URANIUM SERIES RADIONUCLIDES IN THE BIOSPHERE

The U-238 decay chain comprises nine radionuclides with half lives longer than one day, a succession of shorter lived radionuclides, and one stable isotope at the end of the chain, Pb-206 (Figure 1).

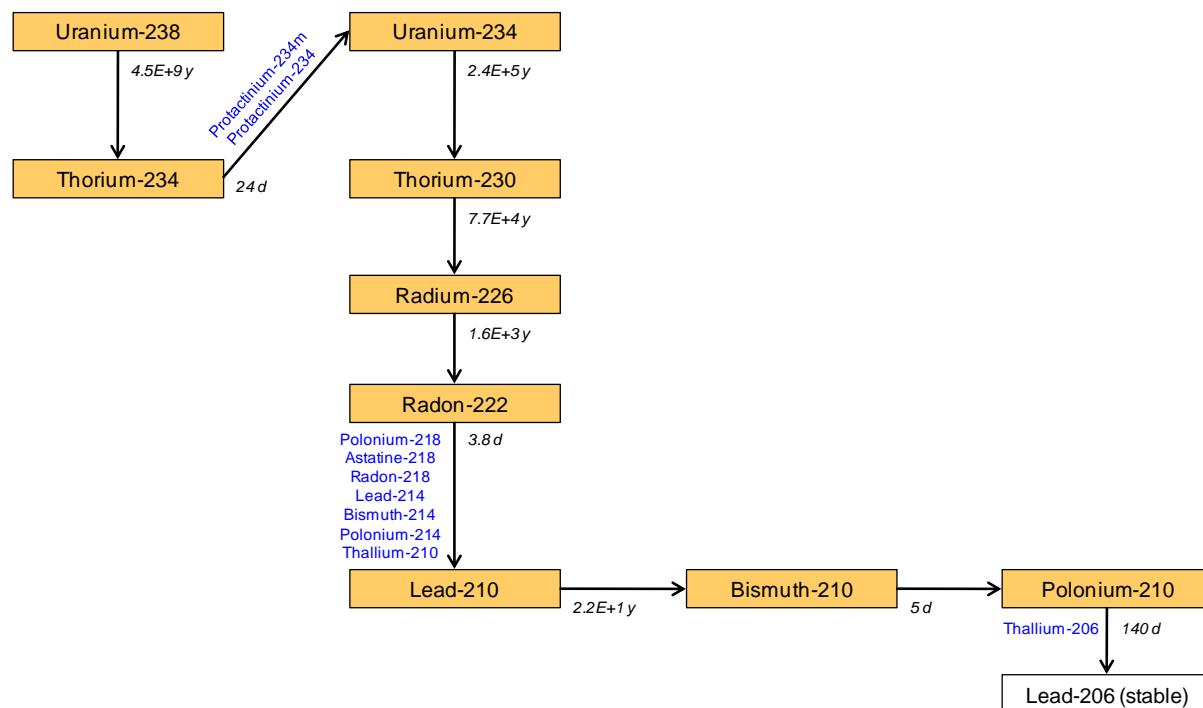


Figure 1 **U-238 Series Decay Chain**

As noted in the introduction, this study is focussed upon the soil-plant interactions within a terrestrial ecosystem. The following sub-sections provide some information on the behaviour of uranium series radionuclides in soil and plants, with a focus on information provided within the recent update to the IAEA TRS 364, as reported in IAEA TRS 472 (IAEA, 2010) and the associated TECDOC 1616 (IAEA, 2009).^a

2.1 BEHAVIOUR OF URANIUM SERIES RADIONUCLIDES IN SOIL

2.1.1 URANIUM

Uranium occurs in valences +3, +4, +5 and +6. In soils, the valences +4 and +6 are the most important. U(IV) dominates at Eh < 200 mV, which is typical for waterlogged to wet soils. U(IV) tends to strongly bind to organic and inorganic matter and to precipitate, and it is, therefore, immobile. The most oxidized state for U in nature is U(VI). At pH below 5, U (VI) is present as the uranyl ion. At a higher pH, the uranyl ion hydrolyzes, forming a number of aqueous hydroxide complexes. The hydrolysed species often dominate U(VI) speciation in the absence of dissolved inorganic ligands such as carbonate, fluoride, sulphate or phosphate. In the presence of dissolved carbonates, U(VI)

^a This following text has been adapted from the February 2010 workshop report (Limer, 2010).

forms strong carbonate complexes. At the pH range of 6 to 10, uranium is largely partitioned into three stable complexes: the acid biphosphate, bicarbonate and tricarbonate. The oxidized uranyl phosphate, sulphate and carbonate complexes are soluble and readily transported. Moreover, at pH < 7 uranyl ions are also bound to dissolved organic matter (DOM), enhancing uranium mobility and possibly changing plant availability (e.g. Reszat and Hendry, 2007; Takeda et al., 2008)

Since so many factors affect K_d (pH, dissolved carbonates, amorphous Fe content, soil mineralogy, CEC, soil organic matter content, DOM), K_d values exhibit large variability, see Sheppard (2011). However, values show a specific trend in relation to the pH. In general, the sorption of uranium in soils is low at pH values less than 3, increases rapidly with increasing pH from 3 to 5, reaching a maximum in the pH range from 5 to 7 and then decreases with increasing pH at pH values greater than 7. One important source of variability in the relationship between K_d value and pH is the heterogeneity in soil chemistry and mineralogy. Soils containing larger percentages of iron oxide minerals and mineral coatings and/or clay minerals will exhibit higher sorption than soils dominated by quartz and feldspar minerals.

The IAEA TECDOC 1616 (IAEA, 2009) recognises the interplay of carbonate as well as pH and Eh in determining U(VI) behaviour. Although many existing assessment models do not account for chemical speciation (e.g. Thorne, 2007), a model developed for CIEMAT uses the water content of a layered soil model as a proxy for redox conditions in order to determine the chemical speciation of a radionuclide (Thorne and Pérez-Sánchez, 2010). It could therefore be questioned as to the relevance of focussing on Eh for U-series radionuclides rather than pH if carbonate is the factor which is dominating in terms of U-speciation; the modelling of such phenomena has been considered previously (e.g. Wheater et al., 2007).

2.1.2 THORIUM AND RADIUM

Thorium is highly retarded in soils (retardation factors of the order 1E+3, with K_d approximately $1\text{ m}^3\text{ kg}^{-1}$), and details of soil characteristics are not particularly important.

Radium occurs in nature as a divalent cation. It has a high affinity for the specific exchange sites of the soil, as shown by the fact that organic matter sorbs about ten times as much radium as clay. It also co-precipitates with barium and strontium to form insoluble sulphates. Due to its alkaline character, it is not easily complexed. There has been limited research on defining K_d values. In geochemical equilibrium models, often data for barium, which may act as an analogue, are recommended for assessing the behaviour of radium. High Ca levels in the soil solution or exchangeable phase and low organic matter and clay content are conducive to higher radium mobility and availability. Overall, IAEA (2009) reported that not enough data and phenomenological comprehension have been assembled so far to mathematically relate K_d values to any of these soil properties.

2.2 UPTAKE BY PLANTS

Although extensive work has been done on U solubility in soils, there is comparatively little information regarding the uptake and translocation of U by plants as affected by soil properties. Generally, it appears that uptake of uranium decreases from sandy through loamy to clay soils. The presence of organic matter generally decreases plant uptake of uranium. There is not much evidence for bioaccumulation of uranium in plants, but there is some evidence for its exclusion. Thorium is known to be bio-excluded; a substantial degree of radium exclusion is not generally accepted but can occur, and is probably dependent on soil mineralogy and geochemistry. Compared with many other radio-

elements, little information exists on the uptake of radium from soil. Among the reported concentration ratios, large discrepancies are noticeable.

Within IAEA TECDOC 1616 (IAEA, 2009), there is a substantial range of variation in the reported soil-plant transfer factors for uranium, with typical differences between the crop groups of a factor of ten, but typical ranges within a crop group of one to five orders of magnitude. For Th and Ra, the observed ranges cover seven orders of magnitude. The majority of data for all the elements were reported without information on soil properties, with only about 50% of the entries containing information on soil type. Information on pH, CEC or organic matter content was even less frequently recorded. The reported values tend to be higher on coarser textured soils, but the values derived by texture class in IAEA (2009) are seldom significantly different.

3. ANALYSIS OF KEY PROCESSES FOR U-238 SERIES RADIONUCLIDES IN A SOIL-PLANT SYSTEM

This section presents an audit of the features, events and processes (FEPs) that would be relevant to modelling U-238 series radionuclide behaviour in the soil-plant system, following release of the relevant radionuclides in groundwater, either via natural discharge to the surface or through well abstraction. The objective is to better justify and transparently explain conceptual models. FEPs are terms used to help to define the relevant scenarios for safety assessment studies, whereby:

- features include the components of the site, such as soil and water bodies;
- events include those incidents that may occur in the future; and,
- processes are related to ongoing actions, natural or human, for example irrigation of agricultural land.

The scenarios need to consider how radionuclides might migrate and disperse or accumulate within the ecosystem, as well as how radiation exposure might arise due to the resulting distribution of radionuclides within and among particular environmental media. In the current project, the focus has been on the migration aspects in a soil-plant system. Consideration is given to a constant soil-plant system, such that there are no external FEPs (such as climatic change) acting upon the system.

The reason for carrying out a FEP audit is as follows. In order to have confidence in a conceptual model used within a safety assessment, e.g. for geological disposal of radioactive waste, it is helpful to demonstrate that all the potentially relevant FEP's have been considered, then present transparent arguments why they have been included or excluded in the assessment model. Even if a FEP is not included in a particular model, the FEP analysis demonstrates that the FEP has been considered in the safety assessment, and has not been ignored. In addition, by auditing a model against independently developed FEP lists and interaction matrices^a, one effectively achieves a peer review of the inclusion of potentially relevant information within the corresponding assessment.

Therefore, in this section, a FEP and interaction matrix analysis is described in two stages:

- In Section 3.1, an interaction matrix for U-238 (U-234 is assumed to behave identically to U-238), Th-230 and Ra-226 in a soil-plant system is given. A summary of those FEPs that are regarded as being potentially important for each radionuclide is also given.
- In Section 3.2, descriptions of those FEPs which are regarded as being potentially important for each radionuclide are given.

This matrix has been developed using the IUR general matrix representation for the migration of radionuclides in a terrestrial ecosystem as a starting point (IUR, 2006; see also Appendix A). The summary of 'key' processes has been used in Section 4 to audit the models that have been considered in this study.

^a The use of interaction matrices in the context of model development for the biosphere is described in detail in IAEA (2003).

3.1 INTERACTION MATRIX FOR U-238, U-234, TH-230 AND RA-226

Since the study is focussed upon the soil-plant system, ‘animals’ have been removed from the IUR general interaction matrix. Soil atmosphere has also been removed as it is unlikely to be a feature relevant to these radionuclides. Vegetation has been included as a single leading diagonal element. However, it should be recognised that the distribution of radionuclides between the above-ground plant compartment and that below ground (i.e. roots) will differ. This will have implications for dose assessment modelling and the cycling of radionuclides between plants and soils, which will vary with crop type and assumptions relating to cropping.

The interaction matrix for these radionuclides is shown in Figure 2. The matrix is focused on those processes relevant for the transfer of radionuclides from groundwater to agricultural plants and soils during the process of irrigation. Those processes considered to be key for all the radionuclides under consideration are shown in **bold text**. For Ra-226 the processes of mineral precipitation and dissolution are considered as being of potential importance if there are high levels of sulphate present (e.g. in locations where phosphogypsum industrial activities occur or where a uranium mine water treatment plant exists); these processes are indicated using underlined text. Where such activities occur, additional processes may warrant consideration (for example, direct transfer of geosphere materials to the biosphere during mining activities and the potential for leaching of radionuclides into soils).

In the matrix in Figure 2, the term ‘suspension’ is used in A8 rather than ‘resuspension’, since the source of the radionuclide is the geosphere rather than the biosphere. With this change of terminology, ‘deposition’ has been added at H1. One new FEP has been added: sediments in the water body interacting with soil organic matter (E2). This is because, in the long term, there is some possibility that such sediments might be used for agricultural purposes.

Several FEPs which appear in the IUR generic matrix (Appendix A) have been removed from Figure 2. Some were removed because they were not relevant to a soil-plant agricultural ecosystem contaminated by radionuclides in groundwater:

- Root uptake from surface water bodies (C2 in the IUR matrix);
- Root exudation, senescence and death in surface water bodies (B3 in the IUR matrix).

Some were removed because they were considered by the project participants as not relevant for the particular radionuclides and system under consideration:

- Photosynthesis (C1 in the IUR matrix) – removed on the grounds that this is not a process relevant to the incorporation of the radionuclides of interest within plant tissues;
- Precipitation, gas sorption (D1 in the IUR matrix) – the radionuclides of interest are not associated with gaseous forms (note the current study does not consider Ra-226 progeny such as Rn-222 gas);
- Diffusive exchange, pressure pumping (both at outcrop; H1 in the IUR matrix) – no gaseous forms of the radionuclide of interest hence no input from the atmosphere;
- Evaporation, gas evolution (A2 in the IUR matrix) – no gas generation anticipated;

- Transpiration, respiration (A3 in the IUR matrix) – no gaseous forms of the radionuclides will be generated within plants so losses to atmosphere by these mechanisms are not anticipated; and
- Evaporation (A4 in the IUR matrix) – radionuclides will remain associated with soil water and soil organic and inorganic matter.

Fertilisation has been kept in the interaction matrix since fertilisation can strongly influence soil chemistry. Liming for example changes in soil pH, and phosphate fertilisation may change the uranium concentration in soil solution.

It may be noted that the consideration of the FEPs above takes little account of the effects of decay and ingrowth. The different soil chemistry of uranium, thorium and radium mean that they are not typically found in secular equilibrium in soils, see Annex B of UNSCEAR (2006). Activity levels in subsoil minerals, which contribute to soil content, as well as activity in groundwater upwelling from below may also give rise to variations, with different processes dominating in different conditions. This complicates the application of natural analogue data (Miller and Smith, 1993; Miller et al, 1997).

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| | A | B | C | D | E | F | G | H | I |
|----------|--|---|--|--|--|---|--------------------------------|--|---|
| 1 | Atmosphere | | 1) Deposition | 1) Deposition | 1) Deposition | | | 1) Deposition | 1) Deposition (at outcrop) |
| 2 | | Groundwater | 1) Diffusion | 1) Irrigation 2) Root uptake | 1) Irrigation 2) Capillary rise | | | | |
| 3 | 1) Droplet production | 1) Leaching | Surface water bodies | | | 1) Sediments | | 1) Sediments | |
| 4 | 1) Pollen and seed release 2) Leaf fall 3) Release of other organic material | | | Vegetation | 1) Root exudation | 1) Litter fall 2) Senescence and death | 1) Symbiotic association | 1) Root exudation | 1) Litterfall (at outcrop) |
| 5 | | 1) Leaching | 1) Throughflow 2) Seepage | 1) Root uptake | Soil solution | 1) Sorption 2) Fixation | 1) Uptake | 1) Sorption 2) Fixation 3) Diffusion 4) Mineral precipitation | 1) Advection 2) Diffusion |
| 6 | 1) Suspension | | | 1) External contamination | 1) Desorption 2) Release during degradation | Soil organic matter | 1) Ingestion 2) Utilisation | 1) Complex formation | 1) Particle transport 2) Colloid transport |
| 7 | 1) Suspension | | | 1) External contamination | 1) Excretion 2) Mineralisation | 1) Death and decomposition 2) Fertilisation | Soil microbiota | 1) Fertilisation | |
| 8 | 1) Suspension | | | 1) External contamination | 1) Desorption 2) Mineral dissolution | 1) Microbial metabolism | 1) Ingestion 2) Utilisation | Soil inorganic matter | 1) Particle transport 2) Colloid transport |
| 9 | 1) Suspension (at outcrop) | 1) Desorption 2) Mineral dissolution | 1) Desorption 2) Mineral dissolution | | 1) Diffusion 2) Colloid transport | | | 1) Chemical & mechanical weathering | Geosphere (below soil) |

Figure 2 Interaction Matrix for Migration and Accumulation of U-238, U-234, Th-230 and Ra-226 in a Soil-Plant System

Table 1 contains a summary of the FEPs that have been identified as being of potential importance for U-238, U-234, Th-230 and Ra-226.

Table 1 Summary of Key FEPs for U, Th and Ra in a Terrestrial Soil-Plant System

| FEP | Radionuclide | | |
|--|-----------------|--------|--------|
| | U-234 and U-238 | Th-230 | Ra-226 |
| Advection | X | X | X |
| Deposition | X | X | X |
| Diffusion | X | X | X |
| Leaching (Groundwater recharge) | X | X | X |
| Irrigation | X | X | X |
| Litter fall | X | X | X |
| Mineral dissolution | | | X |
| Mineral precipitation | | | X |
| Release during degradation | X | X | X |
| Root uptake | X | X | X |
| Seepage | X | X | X |
| Senescence and death | X | X | X |
| Sorption (absorption, adsorption and desorption) | X | X | X |
| Through-flow | X | X | X |

3.2 PROCESS DESCRIPTIONS

Descriptions of the key FEPs identified in Table 1 are given below. These descriptions have been taken from IUR (2006) and checked and modified for relevance to the current project, with a focus on agricultural systems.

Absorption

Sorption and/or adhesion of a layer of ions from an aqueous solution onto a solid surface and subsequent migration into the solid matrix. The process by which atoms, molecules, or ions are taken up from the soil solution and retained on the surfaces of solids by chemical or physical binding.

Adsorption

Whereas absorption is the irreversible binding of molecule to the soil matrix, adsorption of molecules to the soil matrix is reversible.

Advection

Advection is the phenomenon in which dissolved substances are carried along by the movement of fluid displacement. The velocity of fluid displacement depends on the hydraulic conductivity, kinematic porosity and hydraulic gradient of the medium in which the water is present. Seasonal variations in water levels in soil may need to be taken into account. It is noted that capillary rise may also need to be taken into account.

Deposition

The removal of gaseous or particulate material from the atmosphere by precipitation, causing the transfer of material from the atmosphere onto surfaces. Radionuclides can also be deposited directly from the atmosphere as a result of dry deposition of particles. Wet deposition occurs when the radionuclide is captured either by rain or snow during its formation or fall. Occult deposition from mists is included in the wet deposition category. Particles containing radionuclides will generally be

microscopic aerosols of organic or inorganic material, typically derived from soil. However, macroscopic plant parts, e.g. pollen, seeds and leaves, can be vectors of radionuclide transport.

Desorption

The migration of adsorbed entities off the adsorption sites. The inverse of adsorption. Radionuclides may desorb from soil organic matter by reversible processes. However, decay of the organic matter may also result in radionuclide releases that are not characterised as reversible.

Diffusion

Physical process whereby chemical species move under the influence of a concentration gradient. Diffusion is the result of thermal movements in the solution (Brownian motion) and effectively causes a net flux of solutes from a zone of higher concentration to a zone of lower concentration. Diffusion can occur in the pore water or interstitial water of saturated porous media such as soil. Fick's Law is usually applied to the phenomenon and the effective diffusion coefficient for a diffusing contaminant will be controlled by factors such as the concentration gradient, temperature and the tortuosity of the interconnected porosity in the porous medium.

Irrigation

Use of abstracted water to supplement natural supplies to gardens and/or agricultural crops. Irrigation waters containing radionuclides may be applied directly to plants. Strictly, spray irrigation occurs via the atmosphere. However, it may be more convenient to treat it as a direct transfer from water bodies to the external surfaces of plants. Radionuclides present in surface water bodies {or groundwater from wells} may be transferred directly to soil solution in irrigation waters, either applied directly to the surface or by spray techniques. The latter pathway is strictly via the atmosphere, but it may be more convenient to treat it as a direct route of contamination.

Leaching (Groundwater recharge)

The percolation of incident precipitation and other surface waters to groundwater systems resulting in the transfer of dissolved radionuclides from surface waters and/or soil solution to groundwater.

Litter fall

Plant material, such as leaves and twigs falling from plants to the surface soil litter layer. The material then decomposes and can be mixed within the upper soil layer as a result of ploughing or bioturbation processes.

Mineral dissolution and co-dissolution (for Ra-226 in some conditions)

A process by which material in the solid phases is incorporated into the liquid phase. It is affected by local Eh, pH, solubility limits and the presence of other chemical species. It involves the aqueous removal of elements from the crystalline lattice of the mineral surface. This solid-water interaction process is controlled by thermodynamic factors and the water composition, including pH. The dissolution of the different constituent elements may proceed in a congruent fashion that follows the stoichiometry of the lattice structure or in a non-congruent way when certain elements are preferentially dissolved first. Co-dissolution refers to the process whereby co-precipitated trace elements or radionuclides are released from their positions within the lattice during the dissolution process. Once freed these soluble trace constituents may migrate. The dissolution of minerals occurs when rocks suffer chemical weathering. It may also occur when meteoric water percolates down through the unsaturated zone.

Mineral precipitation (mineralisation) and co-precipitation

Involves the assembly of dissolved aqueous elements into a crystalline lattice to form a solid phase. This solid-water interaction process is controlled by thermodynamic factors and the water

composition, including pH. Co-precipitation refers to the process whereby trace elements or radionuclides are incorporated into the lattice structure. A trace element is incorporated either because it can form an ideal solid-solution or because its size and charge are thermodynamically favourable for inclusion. Mineral precipitation may occur during weathering when a new phase may immediately form following mineral dissolution. It may also occur when mineralised water percolates down through the unsaturated zone. Mineral precipitation may affect the net radionuclide retardation capacity of a soil.

Root uptake

The uptake of water and nutrients from soil solution and soil particles by absorption and biological processes within plant roots. Both active and passive uptake can occur, and soil moisture status is a major factor controlling the degree of uptake. Mycorrhizal associations can also be important.

Seepage

Radionuclides present in soil solution may be transferred to surface water bodies by advective flow through the soil with discharge at seepage faces or through spring discharge.

Senescence and death

Ageing and subsequent death of plants (whole or certain tissues such as leaves) resulting in the return of materials to soils during plant/tissue decomposition. Bioturbation processes and/or human activities such as ploughing mix those materials into the upper soil layers. The materials may be rapidly degraded in soil, or they may be either physically or chemically stabilised and only slowly degrade.

Sorption

The removal of an ion or molecule from solution by adsorption and absorption. It is often used when the exact nature of the mechanism of removal is not known. The forces responsible for sorption range from 'physical' interactions (van der Waals' forces) to the formation of specific chemical bonds. Sorption depends both on radionuclide speciation (dependent on valence state, hydrolysis, complexation and soil solution composition) and the solid phase composition and surface characteristics. Desorption kinetics are generally slower than sorption kinetics and there may be instances where sorption is effectively irreversible.

Through-flow

A general term for sub-horizontal flow through soils and such discharge. Alternatively downward seepage/infiltration can occur with radionuclides lost from soil in recharge of groundwater.

4. MODELS FOR CONSIDERATION IN THIS STUDY

This section describes the models used in the quantitative model inter-comparisons (Sections 4.1 to 4.4):

- The CIEMAT soil redox model (Thorne and Pérez-Sánchez, 2010);
- Andra's SAMM model (Miquel and Basso, 2010);
- The NDA RWMD Biosphere Assessment Tool (Thorne, 2007); and
- SYVAC3-CC4, used by NWMO (Gierszewski et al., 2004a).

These particular models were used in this study because they were readily available to project members. Two other models, which are of interest to the group because of the nature of the assessment to which they are applied, are also described (Sections 4.5 and 4.6):

- RESRAD-OFFSITE (Yu et al., 2007); and
- The “Calculation Bases Mining” model (BfS, 2010).

As all of these models are fully documented elsewhere; the purpose of this section is to provide a summary of each model; the technical details of the soil-plant aspects of these models are provided in Appendix B. A summary table is given in Section 4.7 to provide an audit of the processes that are currently considered in the models.

4.1 THE CIEMAT SOIL REDOX MODEL

The CIEMAT model is fully described in Thorne and Pérez-Sánchez (2010). The model uses a simplified modelling approach to soil hydrology to estimate seasonally varying water contents and fluxes in a multi-layer soil model.

Rather than considering capillary rise, the model uses head gradients to determine the upward and downward movement of water. It is anticipated that the overall movement of water would be downwards through the soil, but that seasonally varying upward and downward movements would occur in all the upper layers. At some depth, the water content would stop varying seasonally, but upward and downward fluxes are not limited to the region above this position, i.e. flows propagate to drainage at the base of the model.

Plants are assumed only to take up nutrients and radionuclides from the soil using roots that are above the water table. Different plant types are assumed to have differing root profiles with depth and the activity of those roots is dynamically suppressed below the time varying water table.

The model was originally developed to inform the transport of Se-79 in soils and uptake by plants, taking into account the effects of changing soil water content on sorption, volatilization from soil and root uptake. In the case of U-238 series radionuclides, the volatilisation process is disregarded.

The technical details of the model are given in Appendix B.1.

4.2 ANDRA'S SAMM MODEL

In the framework of the evaluation of the impact of a geological storage of high-, intermediate, and low-level waste (HA- MA-FAVL), the Société de Calcul Mathématique (SCM) developed for ANDRA a multi-layer model of transfer of the radionuclides and chemical toxins (TC) in the soil and soil-groundwater transition zone, SAMM (SCM-Andra-Multilayer Model; Miquel and Basso, 2010).

The model uses a one-dimensional representation of the upper part of the geosphere (Figure 3), including the saturated and unsaturated zones between which is located the geosphere-biosphere interface layer. Further up is the soil layer with a defined root distribution and the presence of plants. Transfer between layers can be advective and/or diffusive in both directions, depending on the surface and soil hydrological balance. The latter may change as a function of depth and time, possibly as a result of an imposed rise of the groundwater level. The transfer of radionuclides and TC is in dissolved form, quantified using the Kd.

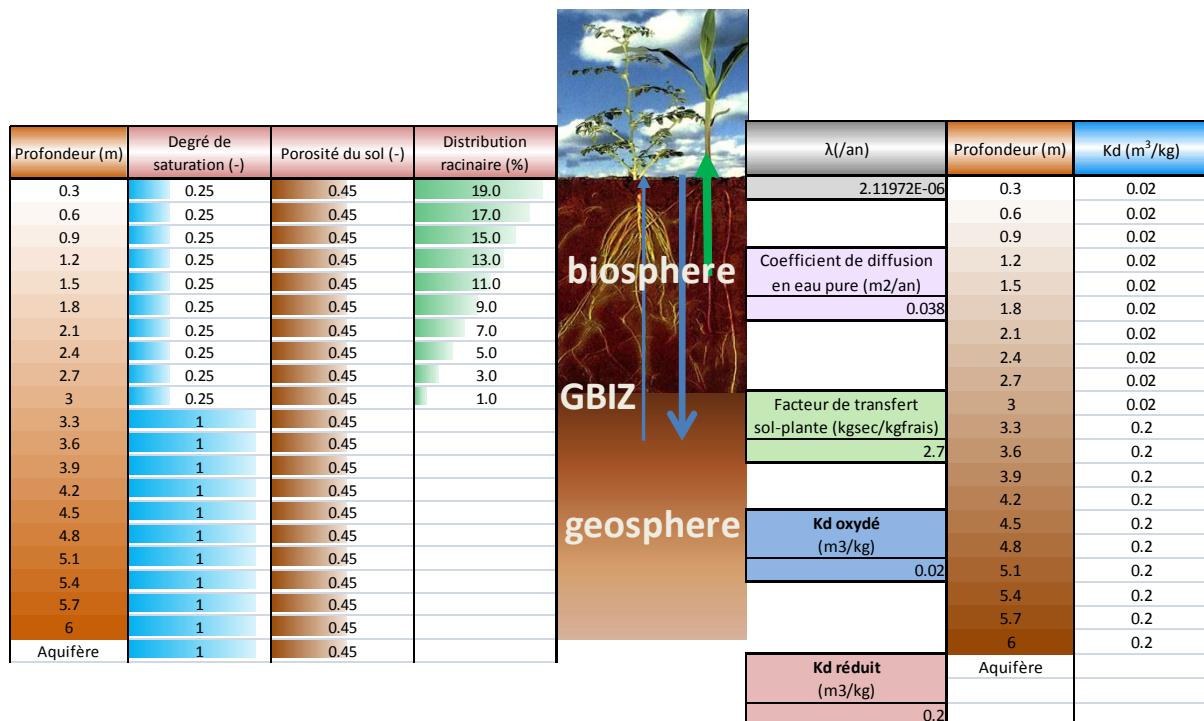


Figure 3 Representation of parts of the set-up for simulations with the SAMM model. Shown in the central part are the biosphere and atmosphere, (redrawn from Marschner, 1995), the permanently saturated geosphere and the geosphere – biosphere interface zone. On the left- and right hand side are shown how Excel is used to discretise the modelled section and how the parameters are defined. The code Matlab used for model development and execution reads these data from Excel.

The technical details of this model are given in Appendix B.2.

4.3 THE NDA RWMD BIOSPHERE ASSESSMENT TOOL

The NDA RWMD has been undertaking a series of generic Disposal System Safety Cases (DSSCs) of a deep geological repository for intermediate- and some low-level radioactive wastes. The latest of these is described in summary in NDA RWMD (2010a). In order to undertake such assessments, it requires a set of radionuclide-dependent biosphere dose factors that relate radionuclide fluxes to the near-surface environment to effective dose rates to representative members of potential exposure groups. Thorne (2007) describes the generic assessment model for the calculation of these biosphere factors as a result of the release of contaminated groundwater, which is assumed to interact with the biosphere either via a natural groundwater discharge, which can also be used for drinking, or via the use of well water for irrigation and drinking. The model is used to calculate the radionuclide concentrations in soil, plants (vegetables and pasture), animal products (meat, milk, liver and kidney from cattle, and fish). The biosphere dose factors take into account exposure from ingestion (of soil, plants, animal products, and water), inhalation, and external exposure. The biosphere status report (NDA RWMD, 2010b) describes NDA RWMD's understanding of the biosphere and the approach adopted to representing the biosphere within the context of the generic DSSC. NDA RWMD's latest consideration of biosphere transport, distribution and radiological impact of uranium series radionuclides released from a deep repository is provided in Thorne and Mitchell (2011).

The model has been implemented in both Excel and GoldSim. The Excel implementation of the model considers the system to be at equilibrium; it is the model with the equilibrium assumption which has been used in this study. The technical details of this model are given in Appendix B.3.

4.4 SYVAC3-CC4

SYVAC is a system model which was developed to address the configuration of a deep geologic repository for used nuclear fuel with in-room emplacement of durable containers, surrounded by dense clay and backfill. The system model is composed of several linked sub-models – the waste form, the containers, the engineered barriers in the vault, the geosphere, and the biosphere. The biosphere model describes the concentration of contaminants in surface water, soils, atmosphere, vegetation and animals, and estimates the consequent radiation dose to a reference person and generic biota. Peak element concentrations in the water, air and soil are also calculated for comparison with chemical toxicity standards.

An earlier version of this model was implemented as a computer code (CC3) and used for the safety assessment submitted to the Canadian Government environment assessment panel in 1994 for the AECL concept for used fuel disposal (Goodwin et al., 1994). An improved model (referred to as PR4, as a “prototype” for CC4) was used for the Second Case Study submitted in 1996 (Goodwin et al., 1996). The system model has continued to be updated and improved. CC4, Version 4.04 was used for a Third Case Study in 2003 (Gierszewski et al., 2004b).

Only a small portion of the biosphere sub-model, the soil model, was used in this study. The soil model is a compartment model capable of interacting with the other regions of the biosphere and geosphere, such as the surface water, atmosphere and groundwater. The technical details of this model are given in Appendix B.4.

4.5 RESRAD-OFFSITE

The RESRAD-OFFSITE code (Yu et al., 2007) is an extension of the original RESRAD code, which was designed for evaluation of radiological doses to an onsite receptor from exposure to RESidual

RADioactive materials in soil (Yu et al. 1993, 2001). Although the “original” RESRAD code is used to model onsite receptors, RESRAD-OFFSITE can model both onsite and offsite receptors.

The methodology and formulations, exposures and scenarios, and parameter values and distributions used in the RESRAD-OFFSITE code are discussed in detail in Yu et al. (2007). Additional modules included in RESRAD-OFFSITE that were not part of RESRAD (onsite) permit modelling of both the transport of radionuclides to locations that are outside the footprint of the primary contamination and their accumulation at those offsite locations (Yu et al., 2007). Models used in the code include the following: a three-dimensional dispersion groundwater flow and radionuclide transport model, a Gaussian plume model for atmospheric dispersion, and a deposition model used to estimate the accumulation of radionuclides in offsite locations and in foods. RESRAD-OFFSITE can be used to model a range of potential exposure pathways and exposure scenarios. Some technical details of this model are given in Appendix B.5.

4.6 “CALCULATION BASES MINING” MODEL (CBM)

Until 2009, Germany had two separate documents (“calculation bases”) for use in the assessment of radiation exposure from environmental radioactivity due to mining, “Calculation Bases for the Determination of Radiation Exposure due to Mining-caused Environmental Radioactivity” and the “Calculation Bases for the Determination of Radiation Exposure due to Inhalation of Radon and its Short-lived Decay Products as a Result of Mining-caused Environmental Radioactivity” (BfS, 2010). These two documents had been produced in the mid-1990s and had been used widely since their publication, due to the fact that these documents were the first to specify calculation models and parameters enabling a “realistic, but sufficiently conservative” assessment of intervention situations in radiological protection according to the German Commission on Radiological Protection (SSK). In particular, these calculation bases were used in remediation projects by Wismut GmbH. In order to account for the natural level of environmental radioactivity involved in measurements, the “Calculation Bases” include consideration of the levels of natural background for all relevant environmental media.

These two documents have since been revised and amalgamated into a single document, “Calculation Bases Mining” (BfS, 2010). This document specifies the procedures to determine mining-caused radiation exposure of members of the public and of workers. They are applicable for the use, decommissioning, remediation, and re-use of mining plants and installations as well as for the use, remediation, and reuse of land contaminated as a result of mining plants and installations. BfS (2010) describes the procedures and parameters to determine effective dose indoors, at underground workplaces, and outdoors, as well as for consumption pathways. The following exposure pathways are considered: external exposure due to gamma-radiation from the soil, exposure due to inhalation of dust, exposure due to inhalation of radon and its short-lived decay products, exposure from ingestion of breast milk and locally produced foodstuffs (drinking water, fish, milk and milk products, meat and meat products, leafy vegetables, other vegetable products), and exposure due to direct ingestion of soil. The technical details of this model are given in Appendix B.6.

4.7 SUMMARY OF PROCESSES CONSIDERED IN THESE MODELS

Table 2 summarises those FEPs relevant to the soil-plant system which are considered in the models in this study. Where a process is considered explicitly in the model this is denoted by “EXP”, and where a process is implicitly considered “imp” is used. Processes which relate directly to the contamination of plants are indicated with a **bold font**.

Most of the models considered in this study include the same processes. None of the models make any implicit or explicit representation of mineral dissolution and precipitation.

Table 2 Summary of FEPs included in the models considered in this study

| Process | Model | | | | | |
|--|--------------|------------|------------------------------------|------------|----------------|------------|
| | CIEMAT model | SAMM | NDA RWMD Biosphere Assessment Tool | SYVAC3-CC4 | RESRAD-OFFSITE | CBM |
| Advection | EXP | EXP | EXP | | | |
| Deposition | | | imp | EXP | EXP | EXP |
| Diffusion | EXP | EXP | EXP | | | |
| Groundwater recharge | EXP | EXP | EXP | EXP | | |
| Irrigation | EXP | EXP | EXP | EXP | EXP | EXP |
| Litter fall | EXP | EXP | imp | Imp | | |
| Mineral dissolution | | | | | | |
| Mineral precipitation | | | | | | |
| Suspension | | | imp | | | |
| Root uptake | EXP | EXP | EXP | EXP | EXP | EXP |
| Seepage | EXP | EXP | EXP | | | |
| Senescence and death | EXP | EXP | imp | imp | | |
| Sorption (absorption, adsorption and desorption) | EXP | EXP | EXP | EXP | | |
| Through-flow (sub-horizontal water flow) | EXP | EXP | imp | | | |
| Capillary rise | imp | EXP | EXP | EXP | | |

5. HYPOTHETICAL SCENARIO DESCRIPTION AND DATA

In this section the hypothetical scenario description is provided, along with the parameterisation of the model applied to it.

5.1 SCENARIO DESCRIPTION

A single source term has been assumed by which radionuclides (U-238, U-234, Th-230 and Ra-226) in groundwater migrate up into the soil profile (i.e. capillary rise and/or diffusion). The subsequent concentration of radionuclides in the soil is to be calculated.

5.1.1 SOURCE TERM

As with the BIOMOVS II hypothetical scenario (BIOMOVS II, 1995), it was assumed the groundwater is contaminated for the first 1000 years, and that after that time is uncontaminated. The following concentrations in groundwater are assumed: 100 Bq L⁻¹ of each of U-238 and U-234 (chemical form U^{IV}), 10 Bq L⁻¹ Th-230 and 200 Bq L⁻¹ of Ra-226.

5.1.2 CLIMATE

The hypothetical scenario assumes the same climate data (Météo France, 2008) as has been used in the BIOPROTA C-14 and Cl-36 studies. It is assumed that any precipitation is free from contamination of radionuclides.

5.1.3 SOIL PROPERTIES

Two one-dimensional columns of soil, each 1 m², are considered, with the following properties (Table 3), consistent with soils in the Pyrenees (Navas et al., 2005).

Table 3 Soil properties for the hypothetical scenario

| Column | Depth (cm) | pH | CO ₃ ²⁻ (%) | OM (%) | Fe oxides (mg / 100g) | Mn oxides (mg / 100g) | Clay (%) | Silt (%) | Sand (%) |
|--------|------------|------|-----------------------------------|--------|-----------------------|-----------------------|----------|----------|----------|
| 1 | 0-5 | 7.81 | 0.97 | 9.2 | 1162 | 40 | 9.2 | 74.4 | 16.4 |
| | 5-10 | 7.84 | 2.0 | 8.2 | 1149 | 38 | 10.5 | 71.1 | 18.4 |
| | 10-15 | 7.96 | 2.9 | 6.4 | 1160 | 39 | 11.9 | 71.8 | 16.3 |
| | 15-20 | 8.07 | 2.5 | 4.1 | 1180 | 38 | 15.3 | 77.0 | 7.7 |
| | 20-25 | 8.30 | 3.2 | 2.9 | 1206 | 39 | 17.1 | 74.5 | 8.4 |
| | 25-30 | 8.24 | 2.6 | 3.1 | 971 | 29 | 15.6 | 75.5 | 8.9 |
| | 30-35 | 8.28 | 2.7 | 2.5 | 1212 | 40 | 15.5 | 70.7 | 13.8 |
| | 35-40 | 8.27 | 2.4 | 2.8 | 1192 | 38 | 16.4 | 76.0 | 7.6 |
| | 40-45 | 8.21 | 2.4 | 2.7 | 1255 | 40 | 15.7 | 75.9 | 8.4 |
| | 45-50 | 8.22 | 2.5 | 2.7 | 1249 | 40 | 18.5 | 79.0 | 2.5 |
| 2 | 0-5 | 7.97 | 12.6 | 9.6 | 886 | 22 | 9.9 | 62.4 | 27.7 |
| | 5-10 | 8.05 | 12.2 | 6.4 | 893 | 22 | 10.3 | 61.9 | 27.8 |
| | 10-15 | 8.19 | 16.2 | 4.8 | 902 | 21 | 11.0 | 68.5 | 20.5 |
| | 15-20 | 8.23 | 8.5 | 4.1 | 928 | 21 | 13.2 | 68.0 | 18.8 |
| | 20-25 | 8.18 | 17.5 | 4.6 | 905 | 20 | 14.4 | 68.4 | 17.2 |
| | 25-30 | 8.26 | 20.3 | 3.0 | 883 | 18 | 16.6 | 70.2 | 13.2 |
| | 30-35 | 8.33 | 19.2 | 3.5 | 862 | 18 | 16.4 | 70.1 | 13.5 |
| | 35-40 | 8.29 | 16.8 | 3.0 | 922 | 22 | 15.2 | 70.7 | 14.1 |
| | 40-45 | 8.26 | 13.7 | 3.4 | 1029 | 28 | 15.7 | 70.3 | 14.0 |
| | 45-50 | 8.23 | 13.2 | 3.4 | 954 | 25 | 15.0 | 69.3 | 15.7 |

5.2 PARAMETER VALUES USED IN THE SAMM MODEL

The parameterisation of the SAMM model is described below.

5.2.1 SITE-SPECIFIC PARAMETERS

A soil profile of 50 cm depth is assumed (consistent with the scenario description), which is subdivided into 10 compartments (Table 4).

Table 4 Discretisation of soil profile in the SAMM model for the hypothetical scenario

| soil parameters | | | |
|-----------------|---------------------------|-------------------|-----------------------|
| Depth (m) | Degrees of saturation (-) | Soil porosity (-) | Root distribution (%) |
| 0.05 | 0.5 | 0.45 | |
| 0.1 | 0.5 | 0.45 | |
| 0.15 | 0.5 | 0.45 | |
| 0.2 | 0.5 | 0.45 | |
| 0.25 | 0.5 | 0.45 | |
| 0.3 | 0.5 | 0.45 | |
| 0.35 | 0.5 | 0.45 | |
| 0.4 | 0.5 | 0.45 | |
| 0.45 | 0.5 | 0.45 | |
| 0.5 | 0.5 | 0.45 | |
| Aquifer | 1 | 0.45 | |

The depth profile goes from the surface down to the saturated aquifer. Homogeneous degrees of saturation and soil porosities have been applied. The soil profile is not considered to be rooted; thus no data is given for the root distribution.

The annual average precipitation over 5 years, as applied in the SAMM model, is given in Table 5.

Table 5 Precipitation Data for the Hypothetical Scenario Used in the SAMM model

| Year | Average precipitation (mm y^{-1}) |
|------|--|
| 1 | 723.1 |
| 2 | 631.9 |
| 3 | 431.0 |
| 4 | 621.9 |
| 5 | 559.9 |

Modelled (evapo-) transpiration is considered constant at 711.4 mm y^{-1} . No irrigation is assumed to occur. The model calculates the hydraulic balance and considers any excess water to percolate down toward the aquifer. In case of a negative hydraulic balance, the required extra water moves up from the aquifer via capillary rise. To test the impact of surface hydrology two cases have been simulated: one case with high and one with low precipitation. Evapotranspiration is constant at 711.4 mm y^{-1} in both cases. In the former case precipitation (723 mm/y consistent with data from Year 1) is higher by 11.7 mm/y compared to evapotranspiration. This surplus of the water balance is considered to

percolate through the soil column. In the latter case a much lower precipitation is considered (431 mm/y, consistent with data from Year 3), a situation which requires 280 mm/y of upward water movement to guarantee a zero hydraulic balance.

5.2.2 RADIONUCLIDE SPECIFIC PARAMETERS

The model considers radionuclide decay and ingrowth. Four members of the U-238 chain are considered, U-238 ($r=1$), U-234 ($r=2$), Th-230 ($r=3$) and Ra-226 ($r=4$). The set-up for the U-238 is shown as an example in Table 6.

The decay constant (λ) is given in per year, the diffusion coefficient in pure water in $m^2 \text{ y}^{-1}$; the soil-to-plant transfer factor is irrelevant as the soil is not modelled as rooted. The K_d (given in $m^3 \text{ kg}^{-1}$) is the Dossier 2005 best estimate for uranium in soils (Andra, 2005). It can change as a function of depth or time, but is set to remain constant for the current study. Critical parameters for the other radionuclides are shown in Table 7.

Table 6 Radionuclide decay and ingrowth in SAMM (U-238 as an example)

| λ (/y) | Depth (m) | K_d (m^3/kg) |
|---|-----------|---------------------------|
| 1.55131E-10 | 0.05 | 0.06 |
| | 0.1 | 0.06 |
| | 0.15 | 0.06 |
| Diffusion coefficient in pure water (m^2/y) | 0.2 | 0.06 |
| 0.038 | 0.25 | 0.06 |
| | 0.3 | 0.06 |
| | 0.35 | 0.06 |
| | 0.4 | 0.06 |
| | 0.45 | 0.06 |
| Soil-to-plant transfer factor ($\text{kg}_{\text{dry}}/\text{kg}_{\text{wet}}$) | 0.5 | 0.06 |
| 0.00082 | Aquifer | |

Table 7 Distribution coefficients assumed in the SAMM model for the hypothetical scenario (Andra, 2005)

| Radionuclide | K_d ($m^3 \text{ kg}^{-1}$) |
|--------------|---------------------------------|
| U-238, U-234 | 0.06 |
| Th-230 | 5.4 |
| Ra-226 | 9 |

The K_d for uranium is the K_d for the oxidised form (U^{VI}). However, in this scenario consideration is given to the less mobile species, U^{IV} . This possible difference has to be kept in mind. Using the lower K_d allows us to compare a more mobile radionuclide (U) with two more strongly sorbed radionuclides (Th and Ra).

5.2.3 AQUIFER CONTAMINATION

In SAMM it is possible to define input parameters as a function of time, allowing conditions to change throughout a calculation; this is possible with all input data. Table 8 demonstrates how the scenario specific groundwater contamination profile has been implemented in SAMM. For the first 1000 years concentrations are constant; between T_1 (1000 y) and T_2 (1010 y) they drop to 0, and then remain at 0 until the end of the simulation (T_3 , 2000 y).

Table 8 Water contamination time series profile for the hypothetical scenario in SAMM

| Time Period | Dates (y) | Duration (y) | Concentration of radionuclide in groundwater (Bq L ⁻¹) | | | |
|-------------|-----------|--------------|--|-------|--------|--------|
| | | | U-238 | U-234 | Th-230 | Ra-226 |
| T_0 | 1000 | - | 100 | 100 | 10 | 200 |
| T_1 | 1000 | 1000 | 100 | 100 | 10 | 200 |
| T_2 | 1010 | 10 | 0 | 0 | 0 | 0 |
| T_3 | 2000 | 990 | 0 | 0 | 0 | 0 |

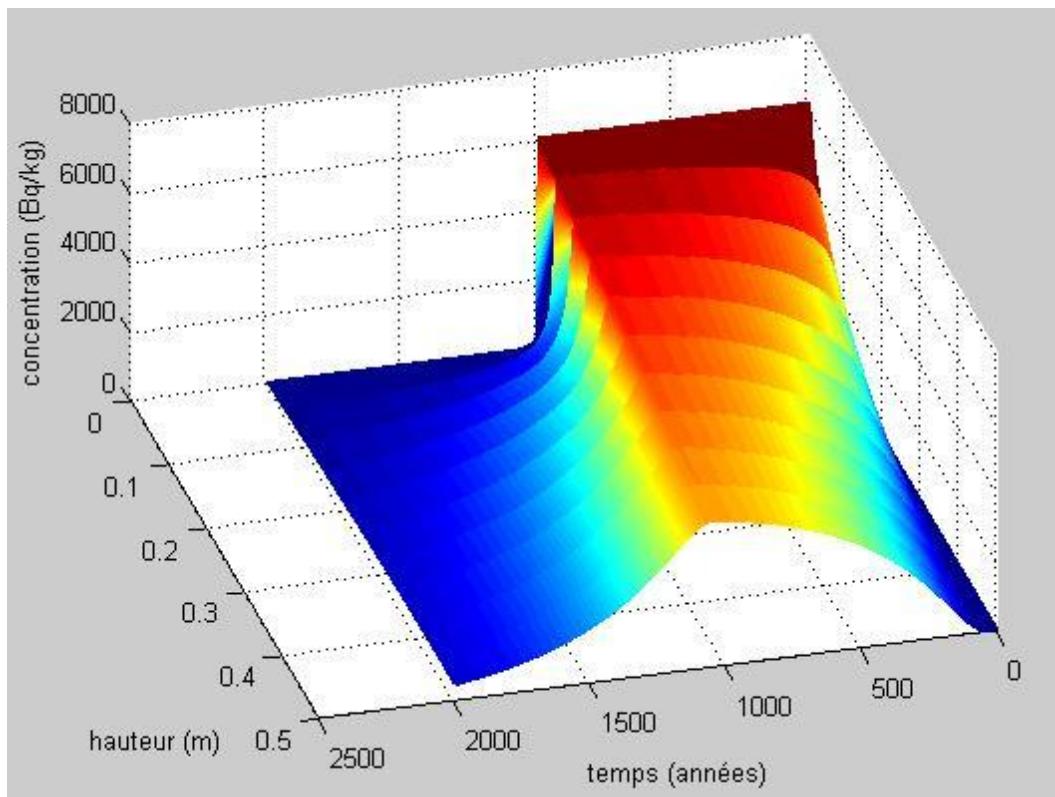
A drop from maximum concentration in an aquifer to 0 within a short period of 10 years may not be very reasonable, particularly for radionuclides with significant sorption in the geosphere. If a significant fraction of a radionuclide is sorbed within the aquifer it will return to solution during a specific period of time depending on advection, diffusion and of course the K_d of the radionuclide in question.

6. HYPOTHETICAL SCENARIO RESULTS

Consideration was given to two cases of upward-moving contaminants: one with high precipitation in which radiation migration will be primarily by diffusion and one with low precipitation leading to capillary rise being the dominant transfer mechanism. These are presented in the following subsections. Only the SAMM model was used for these studies.

6.1 CASE 1: HIGH PRECIPITATION (723 mm y^{-1})

The calculated time-depth distributions of U-238 and U-234 for this case are almost identical (0). The only difference between the two is the decay constant (half lives of 4.5E+9 and 2.5E+5 years, respectively), the impact of which is recognisable for a simulation of 2000 years only in the first decimal place. One can see on the figures at a level (hauteur) of 0 m (i.e. a soil depth of 0.5 m), the interface zone between the aquifer and overlying soil layer. At the input side one can see the constant input peak until 1000 years, a sudden drop during 10 years and then a zero level until the end of the simulation after 2000 years; this saddle-shape result is caused by the input distribution of U assumed in the scenario. Note that the activities given in the figure (and throughout sections dedicated to SAMM) are in total Bq (dissolved and adsorbed) per kg dry soil, which is the concentration that is required to calculate plant concentrations on the basis of soil activities and transfer factors.



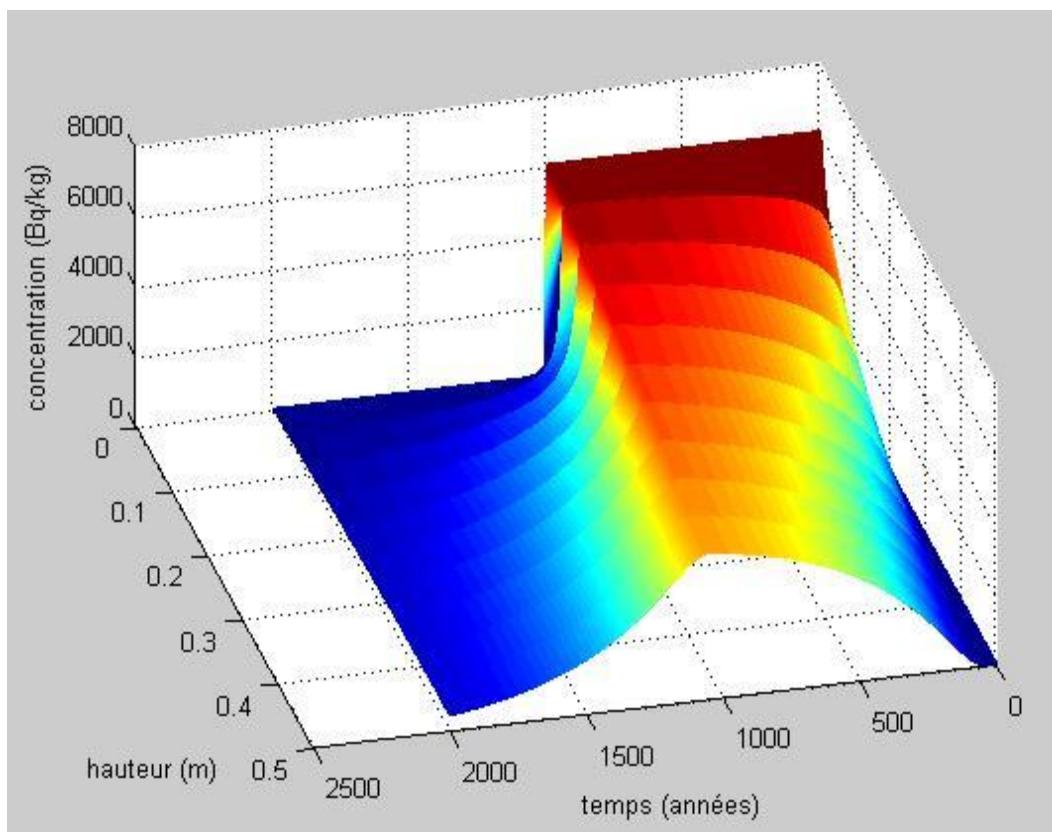


Figure 4 Simulated distribution of U-238 (upper) and U-234 (lower) in soil in the high precipitation case (Bq kg^{-1} wet weight).

The situation is different for Th-230 and Ra-226 because of their higher K_d 's and thus reduced mobility. Both radionuclides diffuse into the soil column, from where only a small fraction moves beyond the next compartment (0.1 m up the soil column); the calculated soil concentrations (in Bq kg^{-1} wet weight) are shown in 0. For the aquifer the total mass-related activities are high because both the dissolved and the sorbed fractions are considered. Since the assumed concentrations of these radionuclides in solution are high (10 Bq L^{-1} and 200 Bq L^{-1} for Th-230 and Ra-226 respectively), and high K_d 's have been assumed, the total concentration in the aquifer becomes high.

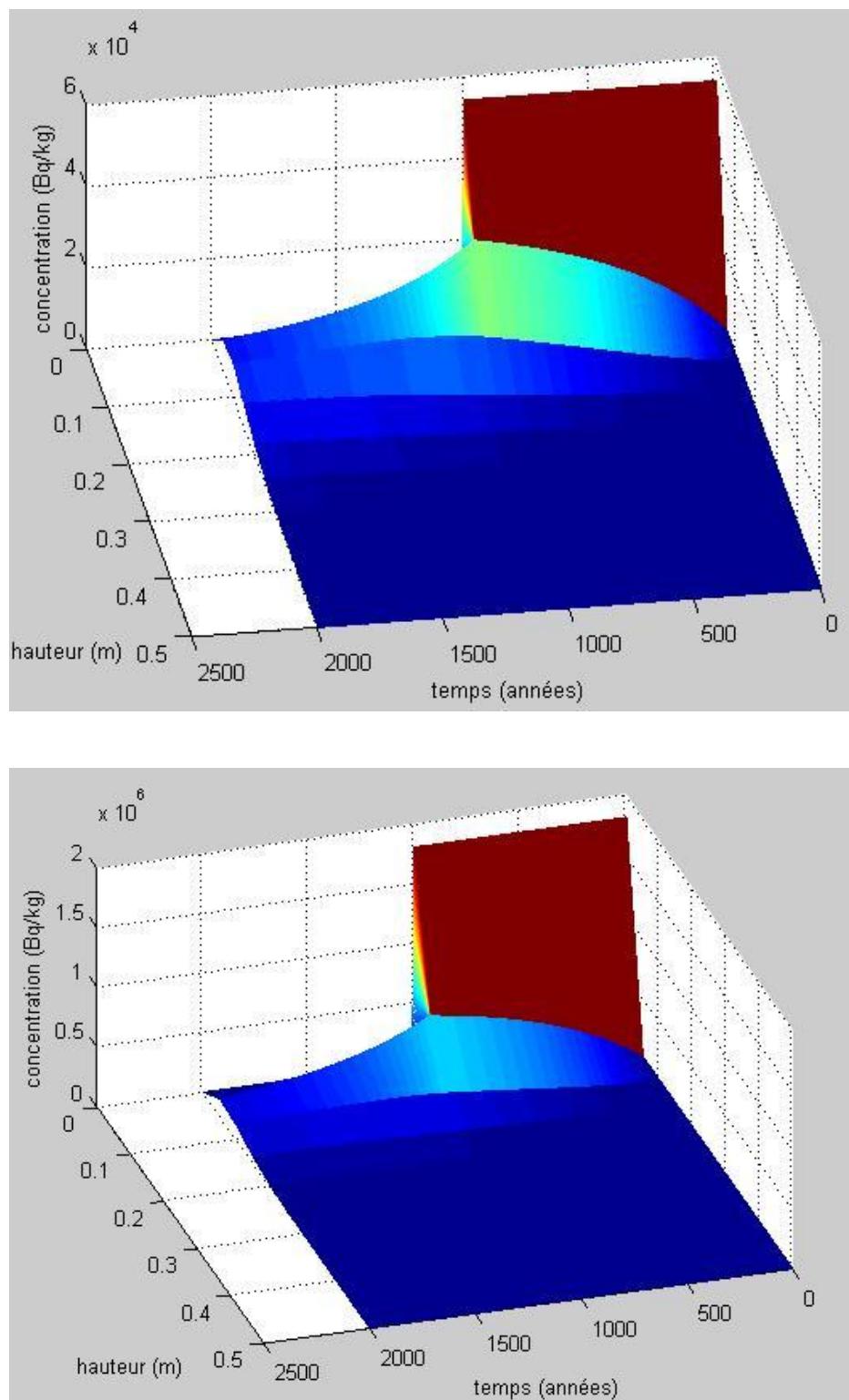


Figure 5 Simulated distribution of Th-230 (upper) and Ra-226 (lower) in soil in the high precipitation case (Bq kg^{-1} wet weight)

The calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 1000 and 2000 years for the high precipitation case are reported in Table 9 and Table 10 respectively.

Table 9 Calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 1000 years for the high precipitation case (Bq kg⁻¹ wet weight)

| Compartment | 1 (near GBIZ) | 2 | 3 | 4 | 5 |
|---------------------------|------------------|-------------|-------------|-------------|-------------------------|
| Upper boundary (m) | 0.05 | 0.1 | 0.15 | 0.2 | 0.25 |
| U-238 | 5.78E+03 | 5.54E+03 | 5.32E+03 | 5.11E+03 | 4.91E+03 |
| U-234 | 5.77E+03 | 5.54E+03 | 5.32E+03 | 5.11E+03 | 4.91E+03 |
| Th-230 | 2.68E+04 | 1.01E+04 | 3.00E+03 | 7.33E+02 | 1.56E+02 |
| Ra-226 | 5.90E+05 | 1.44E+05 | 2.71E+04 | 4.08E+03 | 5.10E+02 |
| Compartment | 6 | 7 | 8 | 9 | 10 (surface) |
| Upper boundary (m) | 0.3 | 0.35 | 0.4 | 0.45 | 0.5 |
| U-238 | 4.73E+03 | 4.56E+03 | 4.41E+03 | 4.27E+03 | 4.14E+03 |
| U-234 | 4.72E+03 | 4.56E+03 | 4.40E+03 | 4.26E+03 | 4.14E+03 |
| Th-230 | 3.45E+01 | 1.23E+01 | 8.40E+00 | 7.50E+00 | 7.20E+00 |
| Ra-226 | 5.43E+01 | 5.00E+00 | 4.30E-01 | 5.00E-02 | 2.00E-02 |

Table 10 Calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 2000 years for the high precipitation case (Bq kg⁻¹ wet weight)

| Compartment | 1 (near GBIZ) | 2 | 3 | 4 | 5 |
|---------------------------|------------------|-------------|-------------|-------------|-------------------------|
| Upper boundary (m) | 0.05 | 0.1 | 0.15 | 0.2 | 0.25 |
| U-238 | 7.73E+01 | 1.51E+02 | 2.18E+02 | 2.79E+02 | 3.31E+02 |
| U-234 | 7.70E+01 | 1.50E+02 | 2.18E+02 | 2.78E+02 | 3.30E+02 |
| Th-230 | 7.06E+03 | 8.14E+03 | 5.46E+03 | 2.68E+03 | 1.06E+03 |
| Ra-226 | 1.44E+05 | 1.18E+05 | 5.27E+04 | 1.67E+04 | 4.18E+03 |
| Compartment | 6 | 7 | 8 | 9 | 10 (surface) |
| Upper boundary (m) | 0.3 | 0.35 | 0.4 | 0.45 | 0.5 |
| U-238 | 3.74E+02 | 4.06E+02 | 4.27E+02 | 4.38E+02 | 4.37E+02 |
| U-234 | 3.72E+02 | 4.05E+02 | 4.26E+02 | 4.36E+02 | 4.35E+02 |
| Th-230 | 3.58E+02 | 1.12E+02 | 3.83E+01 | 1.87E+01 | 1.40E+01 |
| Ra-226 | 8.70E+02 | 1.56E+02 | 2.48E+01 | 3.60E+00 | 6.00E-01 |

It seems surprising that the activity in the lowest section (1 = near GBIZ) dropped more significantly between 1000 and 2000 years of simulation. This is related to the fact that the contamination in the aquifer drops to zero after 1000 years (Table 8) and that from this moment on radionuclides diffuse from the soil section back into the aquifer. It is clear that a fast drop in concentration of adsorbing radionuclides is not very realistic; this should be revised in future simulation exercises.

6.2 CASE 2: LOW PRECIPITATION (431 MM Y⁻¹)

In this case, the precipitation has been decreased from 723 to 431 mm y⁻¹; all other parameters are left unchanged. In this case, in which evapotranspiration is greater than the water supply via precipitation, the extra water required is assumed to move in via advection (i.e. via capillary rise).

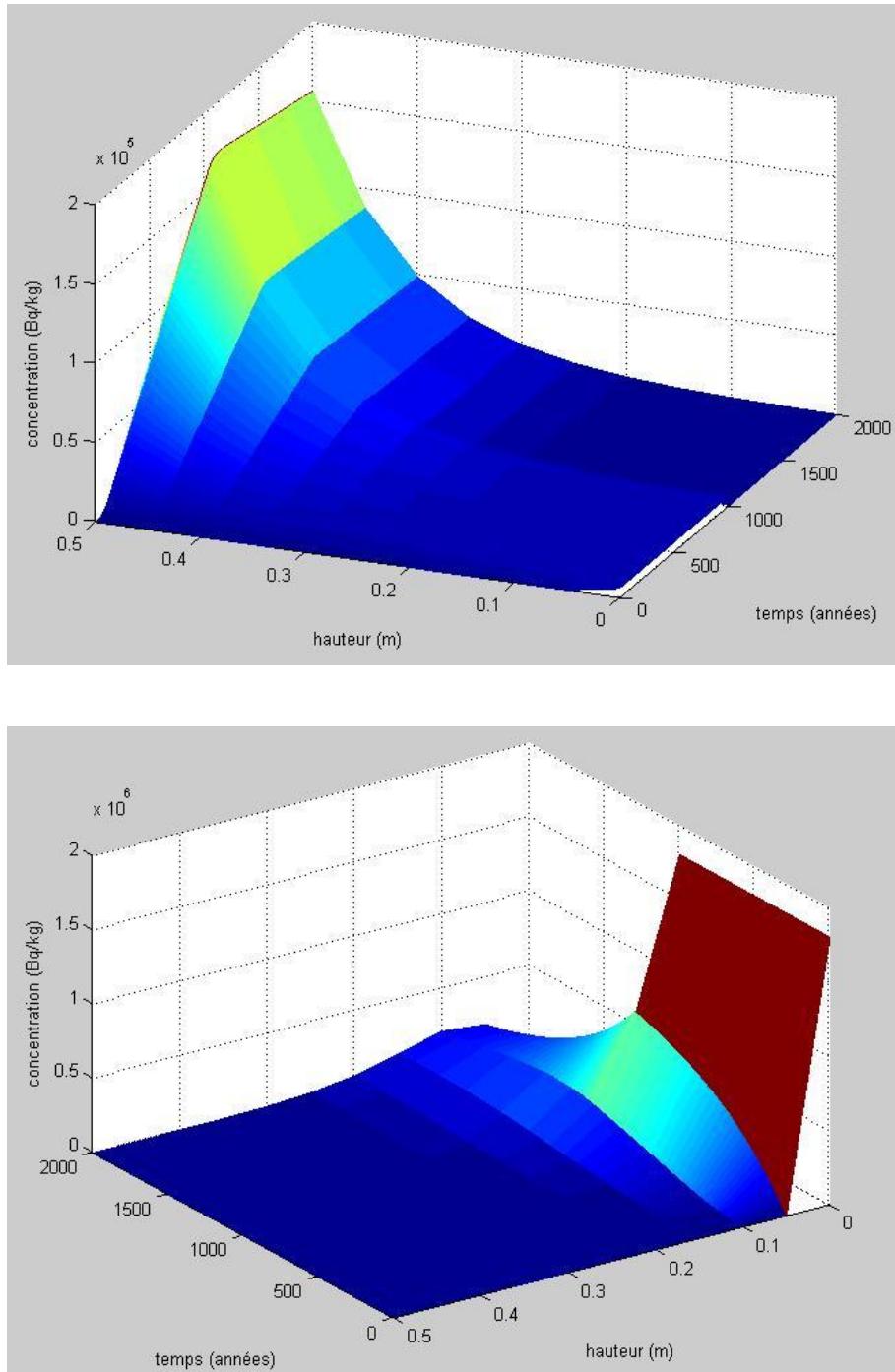


Figure 6 Simulated distribution of U-238 (upper) and Ra-226 (lower) in soil in the low precipitation case (Bq kg⁻¹ wet weight)

In this case, diffusive transport is dominated by advective transport. Further discussion here is focused upon the differences between the high and low precipitation cases, considering that the low precipitation case is dominated by advective upward flux, whereas the high precipitation case is dominated by downward advection with a minor diffusion component.

There is a striking difference for both uranium isotopes between Case 1 (0) and Case 2 (see U-238 in 0, left hand side, representative for both). Because of their low K_d , thus high mobility, uranium is moved upwards by capillary rise (resulting from the low precipitation) and accumulated in the uppermost soil layer in Case 2. The accumulation in the upper parts of the soil profile is so significant that the source term is hardly discernible.

For Ra-226 (0, right hand side) with its high K_d the difference between the high and low precipitation cases is not so significant.

The calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 1000 and 2000 years for the low precipitation case are reported in Table 11 and Table 12 respectively.

Table 11 Calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 1000 years for the low precipitation case (Bq kg^{-1} wet weight)

| Compartment | 1 (near GBIZ) | 2 | 3 | 4 | 5 |
|---------------------------|------------------|-------------|-------------|-------------|-------------------------|
| Upper boundary (m) | 0.05 | 0.1 | 0.15 | 0.2 | 0.25 |
| U-238 | 6.34E+03 | 6.91E+03 | 7.96E+03 | 9.85E+03 | 1.33E+04 |
| U-234 | 6.34E+03 | 6.91E+03 | 7.95E+03 | 9.84E+03 | 1.33E+04 |
| Th-230 | 3.61E+04 | 1.90E+04 | 8.04E+03 | 2.80E+03 | 8.38E+02 |
| Ra-226 | 8.19E+05 | 2.84E+05 | 7.70E+04 | 1.68E+04 | 3.07E+03 |
| Compartment | 6 | 7 | 8 | 9 | 10 (surface) |
| Upper boundary (m) | 0.3 | 0.35 | 0.4 | 0.45 | 0.5 |
| U-238 | 1.95E+04 | 3.07E+04 | 5.12E+04 | 8.82E+04 | 1.55E+05 |
| U-234 | 1.95E+04 | 3.07E+04 | 5.11E+04 | 8.80E+04 | 1.55E+05 |
| Th-230 | 2.36E+02 | 9.05E+01 | 7.97E+01 | 1.20E+02 | 2.06E+02 |
| Ra-226 | 4.77E+02 | 6.46E+01 | 7.90E+00 | 1.10E+00 | 6.00E-01 |

Table 12 Calculated radionuclide concentrations of the four radionuclides throughout the soil profile at 2000 years for the low precipitation case (Bq kg^{-1} wet weight)

| Compartment | 1 (near GBIZ) | 2 | 3 | 4 | 5 |
|---------------------------|------------------|------------|-------------|------------|-------------------------|
| Upper boundary (m) | 0.05 | 0.1 | 0.15 | 0.2 | 0.25 |
| U-238 | 3.81E+02 | 1.06E+03 | 2.27E+03 | 4.42E+03 | 8.26E+03 |
| U-234 | 3.79E+02 | 1.06E+03 | 2.26E+03 | 4.40E+03 | 8.22E+03 |
| Th-230 | 8.02E+03 | 1.30E+04 | 1.25E+04 | 8.74E+03 | 4.95E+03 |
| Ra-226 | 1.77E+05 | 2.07E+05 | 1.34E+05 | 6.17E+04 | 2.24E+04 |
| Compartment | 6 | 7 | 8 | 9 | 10 (surface) |

| Upper boundary (m) | 0.3 | 0.35 | 0.4 | 0.45 | 0.5 |
|---------------------------|------------|-------------|------------|-------------|------------|
| U-238 | 1.51E+04 | 2.73E+04 | 4.89E+04 | 8.75E+04 | 1.56E+05 |
| U-234 | 1.50E+04 | 2.71E+04 | 4.87E+04 | 8.71E+04 | 1.55E+05 |
| Th-230 | 2.40E+03 | 1.09E+03 | 5.68E+02 | 4.94E+02 | 7.10E+02 |
| Ra-226 | 6.77E+03 | 1.77E+03 | 4.10E+02 | 8.73E+01 | 2.27E+01 |

7. LOS RATONES SCENARIO DESCRIPTION AND DATA

7.1 SCENARIO OUTLINE

This scenario is based on real information collected from a former uranium mine site in the south of Spain. The site and environment are described below in terms relevant to uranium series contamination levels and the soil plant system. Given this information, consideration has then been given to assessing the radionuclide concentrations in the soil and crops occurring as a result of migration of the radionuclides up through the soil profile.

7.2 LOS RATONES DESCRIPTION

The Los Ratones former uranium mine is located in the Autonomous Region of Extremadura in the south-west of Spain. The tailings pile associated with the mine covers an area of 2.3 km² (Figure 7). The mine was in use from 1960 to 1974, and restoration work on the tailings pile was undertaken in 1998 and 1999. The site has been subject to environmental sampling and characterisation during the remediation process, and also following the completion of the site remediation process.

Meteorological, chemical, physical, and radiological data have been obtained over a series of discrete sampling campaigns since the site remediation was undertaken. Many of these data have been collected by a team of researchers at the University of Extremadura, funded by Enresa, who have been studying the site (e.g. Vera Tomé et al. 2002, 2003).



Figure 7 Aerial photograph of the mine site before restoration

The mine is located between two villages (Casas de Don Antonio, at a distance of approximately 7 km, and Albalá, at 3 km). There are also farming activities in the zone, and the Maderos River is located to the east and south of the mine site (see Figure 8).

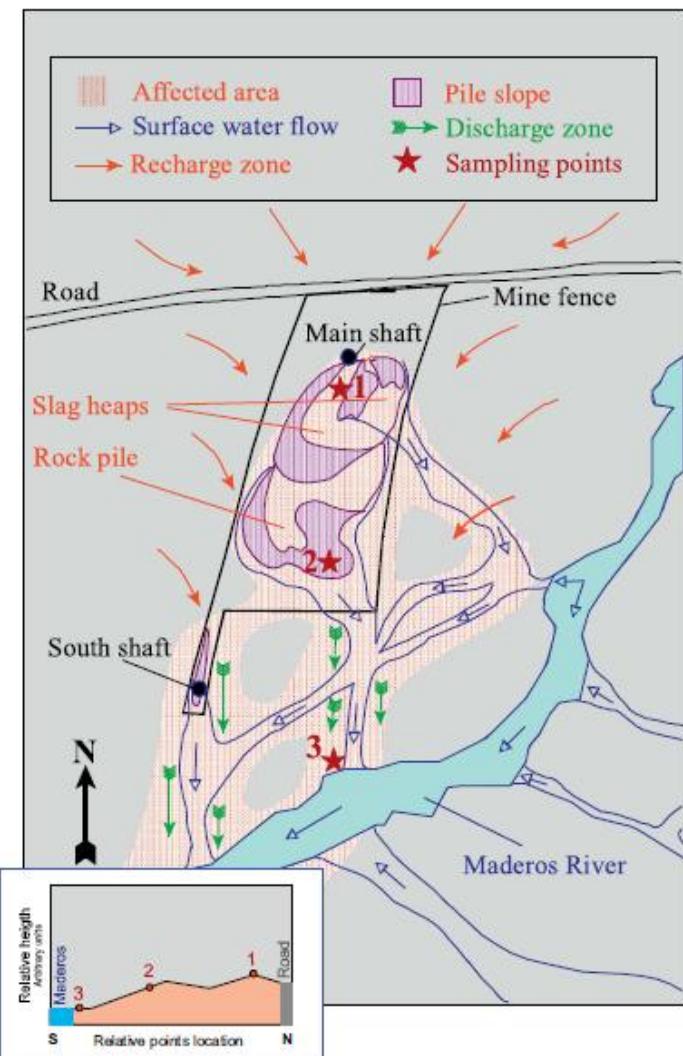


Figure 8 Schematic of the Los Ratones Area

Several spatial and temporal schemes were adopted for the different campaigns of data collection. Daily precipitation and temperature measurements, obtained from a meteorological station situated in the area, were used to derive monthly means as a basis for performing water balance calculations for the whole watershed area. Measurements for characterisation purposes included texture and density of soils and sediments, as well as chemical composition of soils and vegetation samples. Agricultural crops (24% of the total area) present in the area are mainly wheat and barley, with natural grassland (20%) and European oak woodland (56%) occupying the main part of the study area. The hydrology of the watershed, which has a granite underlying geology, is also well understood. Agüero (2005), which contains additional information about the site, describes an assessment model which was developed to assess the behaviour of U-238 series radionuclides in the watershed.

There are two types of location described in relation to Los Ratones – those in areas unaffected by the activities at Los Ratones (denoted by A, B and E in the following subsections) and those in areas downstream of the site (denoted by C and D in the following subsections). The following subsections contain information with respect to climate, soil properties, and the measured activities in the soil, soil-

plant ratios, and distribution coefficients. Uranium activity concentrations as measured in the seasonal Maderos River are also given.

The meteorological data has been used to estimate the position of the water table throughout a year.

Using the information provided below, participants calculated the concentrations of U-238, Th-230 and Ra-226 in soil through a depth profile. The behaviour of these radionuclides, in terms of their concentration, in the soil and crops were calculated over the time period associated with the meteorological data.

7.2.1 CLIMATE

The climatic characteristics of the studied area can be considered to be relatively homogeneous. A representative station at Alcuéscar gives meteorological data (Table 13) from a record of 43 years. The potential evapotranspiration obtained by the Thornthwaite method, for a typical year, is 831.4 mm y^{-1} .

The highest wind frequency is from the west and is characterised by mild and wet air. The mean annual wind velocity is about 2.7 m s^{-1} , with the weak winds (up to 3 m s^{-1}) occurring about 66 % of the time. About 33% of the time moderate to slightly strong (from 3 to 9 m s^{-1}) winds occur and the rest of the time (1%) corresponds to strong or very strong winds (more than 9 m s^{-1}). Spring is the windiest time of the year and winter is frequently calmer. To complement the meteorological data for the site, a file in Excel format was provided, with information from 1983 to 2010 that included the monthly temperature, precipitation, wind speed, average daily insolation in hours and the average percentage of daily insolation compared with the theoretical.

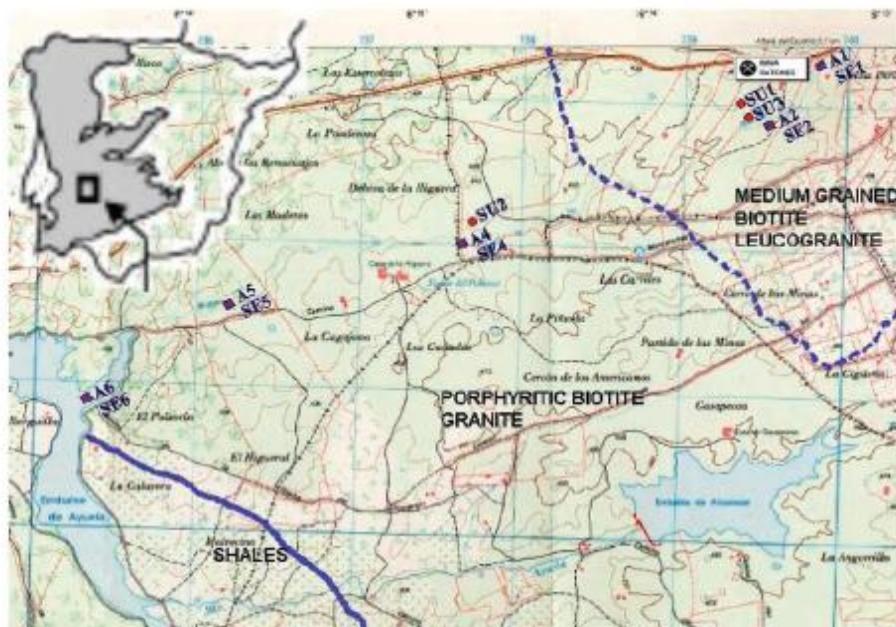
Table 13 Monthly and annual average temperatures, precipitation and evapotranspiration for a 43 year period (1961-2003) from records of the Alcuéscar station (MARM, 2010)

| Month | Temperature ($^{\circ}\text{C}$) | Precipitation (mm) | Potential Evapotranspiration (mm) |
|-------------|------------------------------------|--------------------|-----------------------------------|
| January | 7.4 | 91.4 | 13.7 |
| February | 8.6 | 75.8 | 17.7 |
| March | 11.3 | 55 | 34.1 |
| April | 13.1 | 61.4 | 46.4 |
| May | 17.1 | 47.1 | 80.2 |
| June | 22 | 25.9 | 122.3 |
| July | 25.7 | 5.4 | 160.7 |
| August | 25.6 | 4.7 | 149.5 |
| September | 22.2 | 30.3 | 104.4 |
| October | 16.6 | 74.5 | 60.3. |
| November | 11.2 | 92.3 | 27.3 |
| December | 7.9 | 92.5 | 14.8 |
| Annual mean | 15.7 | 656.5 | 831.4 |

7.2.2 SOIL PROPERTIES

The soil types in the area, at a regional scale, present a broad spectrum from the point of view of their composition, based on the native material. Covering a great part of the granite, poorly developed soils can be distinguished. In the areas close to the dams, agricultural soils are developed, that are irrigated with water from the reservoirs (Figure 9).

Two main soil types can be distinguished on the granite, according to the material on which they are developed and their position with respect to the substratum. Both types belong to the Entisols order from the USDA 1975 classification (Type 1 and 2). Another soil type can be differentiated on the slate material (Type 3).



7.2.2.2 Type 2: Aeric Haplaquent (AQUENT, ENTISOL)

These are poorly evolved soils, developed on alluvial sediments; properties of this soil type are given in Table 15. Sandy ones prevail at the bottom of valleys, with minimal slope and hence slow drainage. These soils have hydromorphic features. They present a superficial mineral horizon sometimes with considerable depth, a substantial proportion of organic matter (OM) and plenty of roots.

These soils are very different to Type 1 soils, having a sudden textural contrast between the two horizons, due to the alluvial contribution being of a different nature. They present high OM content, which implies a high ionic interchange capacity (CEC). They are representative soils of slow draining zones of surface runoff. During wet seasons, temporary flooding can occur.

Morphologically Type 2 soils correspond to lower areas of slopes and the bottom of valleys. The typical profile is maintained through the bottom valley length, except in respect of variations of the A horizon depth.

Table 15 Parameters of the Aeric Haplaquent Soil Characterisation

| Horizon | Thickness (cm) | pH (H ₂ O) | Structure | | | | Organic Matter % | Cation Exchange Capacity meq/100 g |
|---------|----------------|-----------------------|-----------|--------|--------|-------------|------------------|------------------------------------|
| | | | % Clay | % Silt | % Sand | Texture | | |
| A | 7-27 | 4.9 | 24.0 | 40.1 | 35.9 | Loamy | 7.6 | 38.5 |
| C | > 32 | 4.8 | 14.2 | 13.0 | 72.8 | Sandy-Loamy | 1.0 | 6.5 |

7.2.2.3 Type 3: Brown soil, Southern

These soils have variable depth, dominated by those with small depth, and are developed on slate materials. They are moderately acid, with small differences among horizons; properties for this soil type are given in Table 16. They are generally poor in nutritive elements. The Southern ones are developed on plains or smooth slopes; the sub-humid variants have been characterised in areas of higher slope in valleys. Both types show good drainage properties.

The clays in these soils contain illite as a fundamental component and variable quantities of chlorite and kaolinite; this mineralogical composition results in medium values of CEC. This is the soil that is representative of agricultural areas. The slate soils are highly eroded where they have lost the vegetal cover. Due to erosion, sediments originating from these brown soils are produced in smooth depressions, and these are the most fertile ones.

Table 16 Parameters of the Brown Soil characterisation

| Horizon | Thickness (cm) | pH (H ₂ O) | Structure | | | | Organic Matter % | Cation Exchange Capacity meq/100 g |
|---------|----------------|-----------------------|-----------|--------|--------|---------|------------------|------------------------------------|
| | | | % Clay | % Silt | % Sand | Texture | | |
| A | 15 | 6.4 | 13.7 | 38.6 | 40.9 | Loamy | 3.24 | 6.3 |
| B | 35 | 5.8 | 15.5 | 41.2 | 35.9 | Loamy | 0.82 | 9.9 |
| C | > 50 | | | | | | | |

7.2.3 SAMPLING POINTS, PHYSICAL CHARACTERISATION AND RADIONUCLIDE SPECIFIC DATA

Soil samples were chosen considering different substrate types. Two of them, one granite (A; SU4) and another alluvial (B; SU5), were representative of low activity levels, because their location was outside the zone affected by the mining operations (the direction of flow on the surface is NE–SW); sampling location (E; SU1) is another unaffected granitic area, see Figure 10. Another two samples were collected inside the zone assumed to be significantly influenced by the installation, one clearly alluvial (C; SU2), and another (D; SU3) that is a special case, since it is an alluvial soil but with a major contribution from the nearby granitic soils, and is considered as surface runoff alluvial.

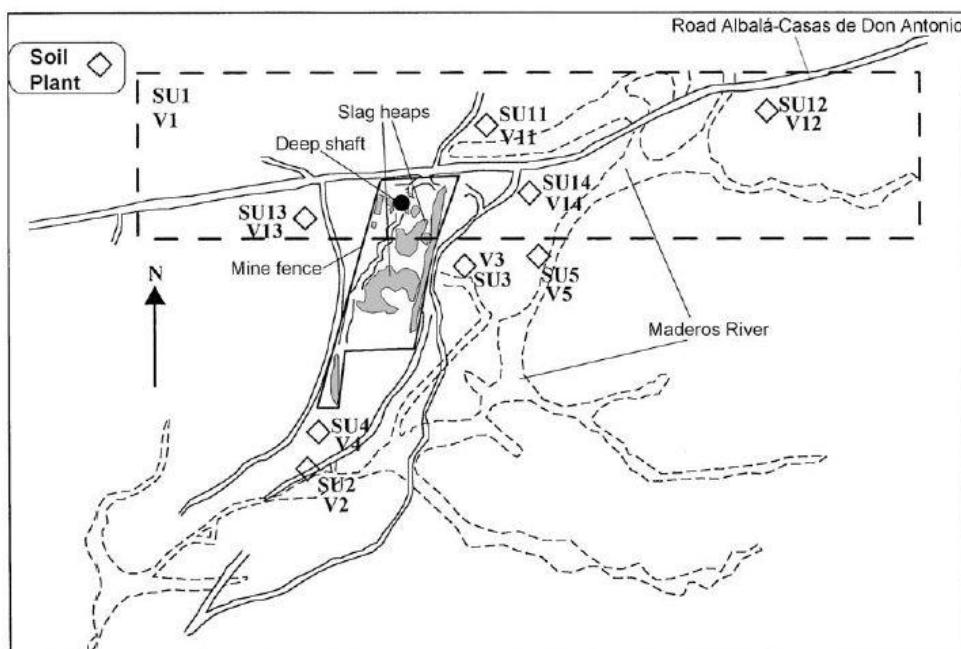


Fig. 1. Sketch of the zone in which the mine is located. The major features and the sampling points are marked. SU1 and V1 denote composite samples of soil and vegetation, respectively, representative of the unaffected granitic area (see text).

Figure 10 Map of the area in which the mine is located. The sampling points for the different soil and plant samples are marked.

In collecting the soil samples, a square frame with an area of 0.25 m^2 was used. In other studies (ENRESA, 1993), the A horizons were determined to be at 5–10 cm, and for soil-to-plant transfer in grass–pasture, other authors consider a depth of 10 cm, hence, a layer of 10 cm depth was also used in the present investigation when collecting the soil samples.

Physical characteristics of the soil samples are provided in Table 17. Radionuclide activity concentrations (means and ranges) are detailed in Table 18.

Table 17 Physical characterisation of the local soil samples (Agüero and Pinedo, 2002 and Vera Tomé et al., 2003)

| Substrate Type | Porfirc Granite | Shales | Alluvial | Muscovite Granite | Alluvial |
|------------------------------------|--------------------|--------|----------|----------------------|----------|
| Sampling Point | E(SU1) | C(SU2) | D(SU3) | A(SU4) | B(SU5) |
| Bulk Density (kg m ⁻³) | 2680 | 2740 | 2650 | 2690 | |
| Texture | Loamy | Loamy | Sandy | Sandy | |
| Water Content (%) | 1.7 | 1.3 | 1.6 | 1.2 | |
| LOI (%) | | 9.1 | 12.1 | 8.5 | 6.2 |

Table 18 Mean values and ranges of the activity concentrations (Bq/kg dry weight) for Uranium and Thorium isotopes and Ra-226 in soil samples according to zone (Vera Tomé et al., 2002)

| Radio-nuclide | E (SU1) * Granitic, OMZ | | C (SU2) Alluvial, IMZ | | D(SU3) Alluvial*, IMZ | | A (SU4) Granitic, OMZ | | B (SU5) Alluvial, OMZ | |
|---------------|----------------------------|---------|--------------------------|------------|--------------------------|------------|--------------------------|---------|--------------------------|---------|
| | Mean | Range | Mean | Range | Mean | Range | Mean | Range | Mean | Range |
| U-238 | 236 | 93-328 | 12073 | 4352-18717 | 9776 | 5190-16000 | 179 | 138-252 | 120 | 113-127 |
| U-234 | 249 | 102-362 | 12103 | 4354-19044 | 9698 | 5138-15739 | 185 | 132-274 | 118 | 116-120 |
| Th-230 | 363 | 225-511 | 10699 | 4730-18779 | 9452 | 3571-18113 | 167 | 119-202 | 150 | 108-195 |
| Ra-226 | 331 | 325-337 | 3529 | 1151-9923 | 7048 | 2609-13420 | 329 | 235-492 | 251 | 191-289 |
| Th-232 | 26 | 14-36 | 30 | 24-37 | 30 | 26-38 | 35 | 29-48 | 24 | 19-30 |
| Th-228 | 37 | 26-56 | 36 | 15-60 | 33 | 28-39 | 52 | 35-97 | 29 | 28-31 |

*With a major contribution from nearby Granitic soils, point E(SU1) is a sample composed of different soil samples taken to the north in an area beyond that subject to the influence of the mine (see Vera Tomé et al. 2002). OMZ and IMZ refer to the sites located either outside or inside the area affected by former mining activity, respectively.

7.2.4 SOIL-PLANT TRANSFER FACTOR AND DISTRIBUTION COEFFICIENTS

Plant sampling points were consistent with those for soils, as described above. The plants were principally grass-pasture (*Fabaceae*, *Poaceae*, *Asteraceae*, etc.). The distribution of these types of plants over the total study area is such that a homogeneous distribution can be considered as a good approximation. The vegetation samples at each point were collected from the surface at which the soil sample was removed. In all cases, only the aerial fraction was sampled. Four sampling campaigns were performed during a one-year period (1998–99) in order to take the variability in meteorological conditions into account.

Three papers have been published on the grass pasture transfer factors: Blanco Rodriguez et al. (2002), Vera Tomé et al. (2002) and Vera Tomé et al. (2003). This last work concluded that the transfer factors they measured were independent of the two substrate types considered, as well as of the two areas of study (an area affected by the mine and an area unaffected by the mine) in which large variations in the concentration of radionuclides belonging to the uranium series were found. The soil-plant transfer factor data (Bq kg⁻¹ dry weight plant per Bq kg⁻¹ dry weight soil) from Vera Tomé et al. (2003) are summarised in Table 19; data were also provided for Th-232 and Th-228 but these radionuclides were not considered in this study.

Distribution coefficients used in the calculations presented in this report are taken from Agüero and Pinedo (2002); see Table 20. Activity concentrations in the surface water are taken from Agüero et al. (2005); see Table 21.

Table 19 Soil-Plant Transfer data for grass-pasture (Vera Tomé et al., 2003)

| Isotopes | Transfer Factor for Each Sampling Point, Bq/kg dw plant per Bq/kg dw soil | | | | All Sampling Points | |
|----------|--|------------------------------------|------------------------------------|-------------------------------------|---------------------|-------------|
| | A (Granitic, Sandy, OMZ) SU4 | B (Alluvial, Loamy, OMZ) SU5 | C (Alluvial, Loamy, IMZ) SU2 | D (Alluvial*, Sandy, IMZ) SU3 | Mean Value | Range |
| U-238 | 0.076 | 0.053 | 0.072 | 0.069 | 0.067 | 0.020–0.250 |
| U-234 | 0.089 | 0.057 | 0.075 | 0.070 | 0.072 | 0.021–0.252 |
| Th-230 | 0.071 | 0.049 | 0.081 | 0.037 | 0.056 | 0.008–0.249 |
| Ra-226 | 0.128 | 0.237 | 0.137 | 0.190 | 0.17 | 0.097–0.504 |

* With major contribution from nearby Granitic soils

Table 20 Soil-Plant transfer data for grass-pasture and distribution coefficients for each sampling points (Agüero and Pinedo, 2002)

| Element | Transfer Factor for Each Sampling Point, Bq/kg dw plant per Bq/kg dw soil | | | | |
|--|---|------------------------------------|------------------------------------|-------------------------------------|-----------------------------------|
| | A (Granitic, Sandy, OMZ) SU4 | B (Alluvial, Loamy, OMZ) SU5 | C (Alluvial, Loamy, IMZ) SU2 | D (Alluvial*, Loamy, IMZ) SU3 | E(Granitic, Sandy, OMZ) SU1 |
| Uranium | 0.0937 | 0.0737 | 0.1038 | 0.1179 | 0.0847 |
| Thorium | 0.2934 | 0.0748 | 0.1473 | 0.0464 | 0.1623 |
| Radium | 0.1300 | 0.2602 | 0.1414 | 0.2780 | 0.1333 |
| Distribution Coefficients for Each Sampling Point ($\text{m}^3 \text{ kg}^{-1}$) | | | | | |
| Uranium | 0.02694 | 0.1060 | 0.08934 | 0.08383 | 0.02834 |
| Thorium | 0.01523 | 0.1052 | 0.07502 | 0.1337 | 0.02048 |
| Radium | 0.02288 | 0.05644 | 0.07657 | 0.05461 | 0.02259 |

* With major contribution from nearby Granitic soils

Table 21 Activity Concentration in Surface waters from Maderos River (Agüero et al. 2005)

| Location | Sampling Campaign | Water Activity Concentration of Radionuclides (mBq L^{-1}) | | |
|------------------------------|-------------------|---|-------|-------|
| | | U-235 | U-238 | U-234 |
| Upstream Sampling Point A1 | 1 st | 0.68 | 16 | 19 |
| Downstream Sampling Point A4 | 1 st | 17 | 400 | 410 |
| | 2 nd | 44 | 750 | 720 |
| | 3 rd | 14 | 260 | 420 |
| | 4 th | 6.8 | 140 | 150 |

7.3 PARAMETER VALUES

In this section details are given on the specific parameterisation of each of the models applied to the Los Ratones scenario.

7.3.1 SAMM

The employed climate scenario is based on average yearly precipitation and evapotranspiration data detailed in Table 13. The calculated evapotranspiration of 1.795 m y^{-1} (Table 22) is considerably higher than the annual mean given in the Los Ratones scenario description (Table 13). The difference is expected to be related to the choice of the model used for the calculation of evapotranspiration (= potential evapotranspiration) (Thornthwaite vs. Turc).^a

An additional parameter, called the average coefficient of effective infiltration is given in Table 22. This parameter is specific to SAMM. The model does not evaluate water flow on the basis of water content, hydraulic conductivity and a physical transport relationship as do classical soil physics models (i.e. Hillel (1998); an empirical parameter is required to scale the fraction of precipitation and upward flux that actually travels within the soil column. It is independent of the amount of evapotranspiration. If the parameter is set to 0, no advective flow is considered, if it is equal to 1 all precipitation and upward flowing water is considered to travel.

Table 22 Average climate data used in all simulation periods

| Parameter | Units | Value Assumed During a Given Time Period | | |
|---|-------------------|--|-------------|-------------|
| | | $T_0 - T_1$ | $T_1 - T_2$ | $T_2 - T_3$ |
| Precipitation | m y^{-1} | 0.6565 | 0.6565 | 0.6565 |
| Evapotranspiration | m y^{-1} | 1.795 | 1.795 | 1.795 |
| Irrigation | m y^{-1} | 0 | 0 | 0 |
| Average coefficient of effective infiltration | - | 0.3 | 0.3 | 0.3 |

No Irrigation is assumed to occur during the entire simulation period. The model calculates the hydraulic balance and considers any excess water to percolate down toward the aquifer. In the case of a negative hydraulic balance (this case), the required extra water moves up from the aquifer via capillary rise or other processes.

Rather than using the initial soil contamination data provided in the scenario description (Table 18), SAMM simulates an initial contamination to have occurred via contaminated ascending groundwater during a default period of 20 years (19.5, Figure 11). The groundwater contamination is calculated according to the present soil contamination. To simplify consideration of the different degrees of soil contamination, an arbitrary distinction was made between low contamination sites (E, A, and B; Table 18) and high contamination sites (C, D; see also Table 23). After this period of contamination, a short period of 1 year is considered during which contamination drops to the value measured in today's surface water, at which it remains constant during the final period of simulation lasting until 40 years for the low contamination case and 47 years for the highly contaminated soils (Figure 11).

^a Andra considered a value of 0.831 m y^{-1} , too low for a (semi-) arid climate.

| Echelle de temps | | T1 | T2 | T3 |
|---------------------|--|------------|------------|------------|
| Dates (années) | | scénario 1 | scénario 2 | scénario 3 |
| | | 19.5 | 20.5 | 40 |
| Nombre de scénarios | | | 3 | |
| Nombre de cycles | | | 1 | |

Figure 11 Definition of the number of scenarios and their duration (years) for the low contamination soils ($T_3 = 47$ for the high contamination case)

As shown later, this “work around” is indeed relatively effective. Newer versions of SAMM (currently in development) will allow definition of initial concentrations and will not require this type of procedure.

Concentrations of the four members of the U-238 decay chain U-238 are roughly at secular equilibrium in the various contaminated soils (Table 18). For this reason, identical groundwater concentrations are assumed for all four radionuclides. The assumed concentrations with time for the two cases considered are given in Table 23.

Table 23 Assumed concentration of the radionuclides in groundwater as a function of time for the low and high concentration soil locations

| Type of soil contamination | Assumed Concentration in Aquifer Water (Bq m^{-3}) | | | |
|----------------------------|---|---------|-------|-------|
| | T_0 | T_1 | T_2 | T_3 |
| Low | 10,000 | 10,000 | 387.5 | 387.5 |
| High | 450,000 | 450,000 | 387.5 | 387.5 |

For the initial soil contamination scenario, identical K_d 's of $0.016 \text{ m}^3 \text{ kg}^{-1}$ were used for all radionuclides at all depths. These were used to give the soil some initial contamination. Site-specific data for K_d and soil-to-plant transfer factors (TF) were considered to apply only to the subsequent inter-comparison scenarios. They have been extracted from Table 18 considering the distinction between high and low contamination soils. An example of how radionuclide specific parameters are defined for application in SAMM is given in Table 24.

Table 24 Definition of site and radionuclide specific parameters for U-238; the soil surface is on top of compartment 0.05 and the aquifer below compartment 1

| λ (/y) | Depth (m) | Kd (m^3/kg) |
|---|-----------|-----------------|
| 1.55131E-10 | 0.05 | 0.538 |
| | 0.1 | 0.538 |
| | 0.15 | 0.538 |
| Diffusion coefficient in pure water (m^2/y) | 0.2 | 0.538 |
| | 0.25 | 0.538 |
| 0.038 | 0.3 | 0.538 |
| | 0.35 | 0.538 |
| | 0.4 | 0.538 |
| | 0.45 | 0.538 |
| Soil-to-plant transfer factor (kg_{dry}/kg_{wet}) | 0.5 | 0.538 |
| | 0.55 | 0.538 |
| 0.000753 | 0.6 | 0.538 |
| | 0.65 | 0.538 |
| | 0.7 | 0.538 |
| Kd (m^3/kg) | 0.75 | 0.538 |
| | 0.8 | 0.538 |
| 0.06 | 0.85 | 0.538 |
| | 0.9 | 0.538 |
| | 0.95 | 0.538 |
| | 1 | 0.538 |

The decay constant (λ) is given in per year, the diffusion coefficient in pure water in $m^2 y^{-1}$, the soil-to-plant transfer factor (TF) in $kg_{dry} \text{ weight plant } kg_{fresh \text{ weight soil}}^{-1}$ and the K_d in $m^3 kg^{-1}$. The K_d can vary as a function of depth or time, but is allowed to remain constant for this exercise. K_d and TF for all radionuclides which were used in the analysis presented in the report are shown in Table 25.

Table 25 Parameter values for the four radionuclides considered in the simulation exercise for the low and high concentration soils (derived from Table 18).

| Radionuclide | K_d ($m^3 kg^{-1}$) | | TF ($kg \text{ dry weight plant } kg_{fresh \text{ weight soil}}^{-1}$) | |
|----------------------------|-------------------------|--------|---|--------|
| Type of soil contamination | Low | High | Low | High |
| U-238, U-234 | 0.0538 | 0.0866 | 0.0753 | 0.0846 |
| Th-230 | 0.0470 | 0.1044 | 0.1301 | 0.0779 |
| Ra-226 | 0.0340 | 0.0656 | 0.1777 | 0.1866 |

It is important to note that plants are not considered to be harvested; thus no plant export is taken into account in the model runs.

7.3.2 NDA RWMD BIOSPHERE TOOL

In the NDA RWMD Biosphere Tool calculations the following assumptions were made:

- The dry soil bulk densities were assumed to be 0.5 times that of the bulk densities reported in Table 17;
- The U-238 and U-234 water concentrations were taken from Table 21, the Th-230 concentration from Lozano et al., (2002) and the Ra-226 concentration from Lozano et al. (2000).

All other data were consistent with the scenario description.

7.3.3 NWMO SYVAC3-CC4

Parameter values for deposition, capillary rise and irrigation are shown in Table 26. For the capillary rise, instead of using the data relating to the concentration of radionuclides measured in the Maderos River (Table 21), the groundwater concentration was calculated from the soil concentrations given in Table 18. In order to convert those data into a groundwater concentration suitable for use in the model calculations, the following equation was used:

$$C_{gw}^i = \frac{C_{soil}^{i,ref}}{K_d \rho_w} \left(\frac{V_{ds}}{V_{gw}} \right) \quad (1)$$

Where:

$C_{soil}^{i,ref}$ is the reference soil concentration (Table 18) [Bq kg⁻¹_{drysoil}]

K_d is the sorption coefficient [m³_{drysoil} kg⁻¹_{drysoil}]

ρ_w is the density of water [kg_{water} m⁻³_{water}]

$\left(\frac{V_{ds}}{V_{gw}} \right)$ is the ratio of the dry soil volume to the groundwater volume in the soil [m³_{drysoil} m⁻³_{water}]. It can be expressed as a function of the water content (θ_s), using the following relationship:

$$(V_{ds} + V_{gw}) \theta_s = V_{gw}$$

The flow of water from the water table in deeper soils to the surface soil compartments is calculated from a simple capillary rise formula for the NWMO model. No information regarding the hydrogeology of the site is given in Section 7.1, thus, nominal NWMO values were used (e.g. depth to water table).

Table 26 Parameters used in SYVAC3-CC4

| Parameter | Description | Units | Value | Source |
|----------------------|--|---|--|--|
| A_F | Area of the field for atmospheric deposition and irrigation | m^2 | 460 000 | 1/5 th of 2.3 km^2 area of mine site. |
| V_d | Dry deposition velocity | m s^{-1} | 0.006 | Garisto et al. (2011) |
| P_{tot} | Total annual precipitation rate | m y^{-1} | 0.656 | Pérez-Sánchez (2010) |
| W_r | Washout ratio | - | 630000 | Garisto et al. (2011) |
| A_{ADL} | Atmospheric particle (aerosol) load from aquatic sources | $\text{m}^3_{\text{water}} \text{ m}^{-3}_{\text{air}}$ | 2.90E-10 | Garisto et al. (2011) |
| C_L^i | Concentration of nuclide i in the water | $\text{mol m}^{-3}_{\text{water}}$ | See Source | U238, U234 and Th230 values from Table 21 Ra-226 values correspond to concentrations at locations M4 and M3 from Lozano et al. (2000) for low and high concentration areas respectively. |
| C_{gw}^d | Groundwater pore water nuclide concentration | $\text{mol m}^{-3}_{\text{water}}$ | See Equation 1 | Pérez-Sánchez (2010) |
| $C_{irr}^f(t)$ | Nuclide concentration of irrigation water at time t for sample point f | mol m^{-3} | See Source | U238, U234 and Th230 values from Table 21 Ra-226 values correspond to concentrations at locations M4 and M3 from Lozano et al. (2000) for low and high concentration areas respectively. |
| Q_{swd} | Summer water deficit | $\text{m}^3_{\text{water}} \text{ m}^{-2}_{\text{soil}} \text{ y}^{-1}$ | 0.175 | Corresponds to PET-P from Table 13 |
| Z_{ss} | Depth of the surface soil layer | m | 0.1 | Table 14 and Table 15 |
| Z_d | Depth to the water table | m | 1.5 | Assumed |
| b | 3 for a fine sandy loam | - | 3 | Garisto et al. (2011) |
| I_{irr}^f | Rate of irrigation for each soil type | $\text{m}^3_{\text{water}} \text{ m}^{-2}_{\text{soil}} \text{ y}^{-1}$ | Sample points A, B and D: 0.1 (sand) Sample Points C and E: 0.06 (loam) | Garisto et al. (2011) |
| $CR^{i,f}$ | Plant/soil concentration ratio for nuclide i and sampling point f | $\text{kg}_{\text{wetbio}} \text{ kg}^{-1}_{\text{wetsoil}}$ | See Source | Table 20 |
| ρ_{soil} | Dry soil bulk density | $\text{kg m}^{-3}_{\text{drysoil}}$ | A | 1345 |
| | | | B | 1325 |
| | | | C | 1370 |
| | | | D | 1325 |
| | | | E | 1340 |
| | | | Table 17. Assume 50 % porosity, such that dry bulk density is halved. | |

| | | | | |
|------------|--|--|------------|--|
| f_{le} | Input leaching rate fraction | - | 0.1 | Garisto et al. (2011) |
| R_T | Average watershed runoff rate | $\text{m}^3 \text{water} \text{ m}^{-2} \text{ soil} \text{ y}^{-1}$ | 0 | Conservatively set to 0 because potential evapotranspiration is larger than precipitation, and no reliable method was available for determining actual evapotranspiration. |
| θ_s | Volumetric water-content of the surface soil | $\text{m}^3 \text{water} \text{ m}^{-3} \text{ soil}$ | A | 0.097 |
| | | | B | 0.067 |
| | | | C | 0.104 |
| | | | D | 0.137 |
| | | | E | 0.017 |
| K_d^i | Sorption coefficient of nuclide i in each sampling field f | $\text{m}^3 \text{kg}^{-1}$ | See Source | Table 20 |

8. LOS RATONES SCENARIO RESULTS

It is noted first that SAMM was used to assess concentrations of the relevant radionuclides in the surface soil at the start of the simulation period, whereas the other models were used to assess concentrations in the surface arising as a result of migration to the surface after the start of the simulation period.

8.1 SAMM MODEL

Low and high soil contamination model cases have been carried out separately, results are summarised independently.

8.1.1 LOW CONCENTRATION SOIL (UPSTREAM SITES)

The first period of simulation from 0 to 19.5 years was required to initialise contamination of the soil column. Figure 12 demonstrates the evolution of the calculated concentration of U-238 through the soil profile during that period. It is sufficient to give only the example of U-238 because contamination is identical for the three other radionuclides (Table 23), and because they have comparable K_d 's and transfer factors (Table 25).

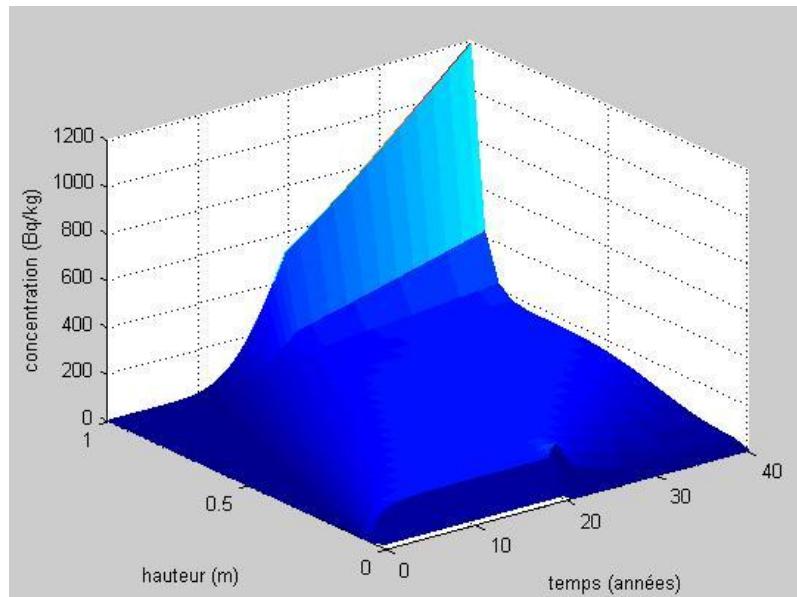


Figure 12 Modelled concentration of U-238 in the low concentration soil as a function of time (tempo, y) and depth (hauteur, m, with 1 = soil surface). The 20 years of intercomparison begin at the date 20 years (tempo – années).

Because of the high flux of ascending water and the low K_d of $0.016 \text{ m}^3 \text{ kg}^{-1}$ for U-238 (and the other three radionuclides) a quasi-equilibrium with a maximum at the surface is established in the soil column after this initial period (Figure 12). The concentrations of all four radionuclides are indeed comparable to what is actually measured in the soil (Table 18). It is from this point on that plant growth is considered and that parameters are derived from the site information.

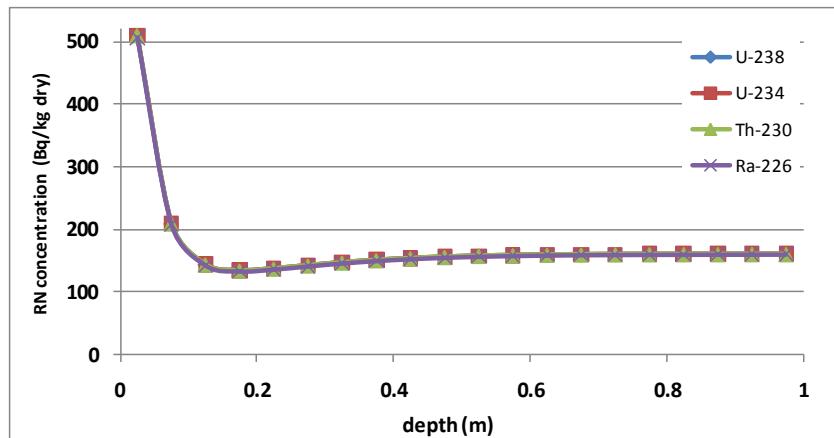


Figure 13 Distribution of the four radionuclides in the soil profile after 19.5 years of initial contamination (0 = surface); giving the onset for the inter-comparison part.

The 3-dimensional representation of the soil concentration (Figure 12) can again be used for observation of the main phenomena. The concentration at depth starts to decrease after 19.5 years to reach “today’s” groundwater (river) contamination of 0.3875 Bq L^{-1} (Table 21). The ascending water as well as root uptake and transfer to the surface cause the general upward movement of all radionuclides with time combined with the slow reduction of concentration with depth. As plants are not considered to be exported from the site, radionuclides taken up by plants are recycled into the first compartment of 0.05 m depth. The concentration with depth at the end of the simulation period shows the significant increase near the surface and the reduction at depth (Figure 14).

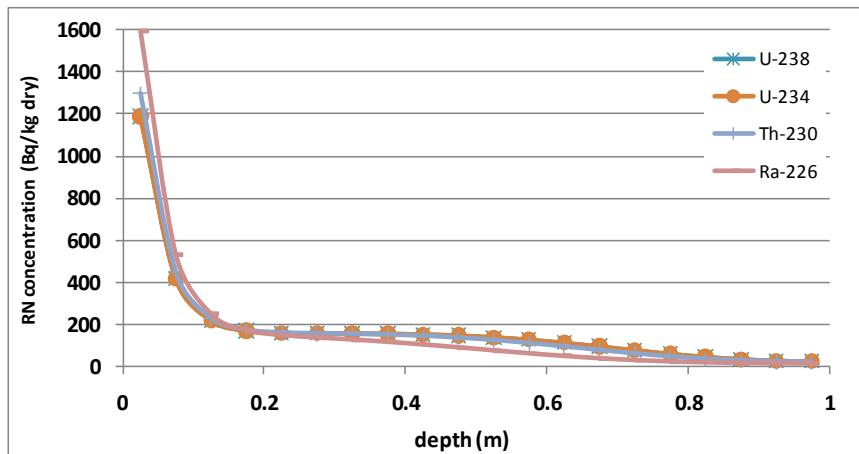


Figure 14 Distribution of the four radionuclides in the soil profile after 40 years of simulation (= 20 years after initial contamination); showing the situation after a period comparable to 1990 through 2010

Finally, Figure 15 presents the results specifically demanded for the inter-comparison exercise, i.e. the concentration of all four radionuclides in the surface layer (0.3 m thickness) and in the plants as a function of time for the period of 1990 to 2010^{Error! Bookmark not defined.}.

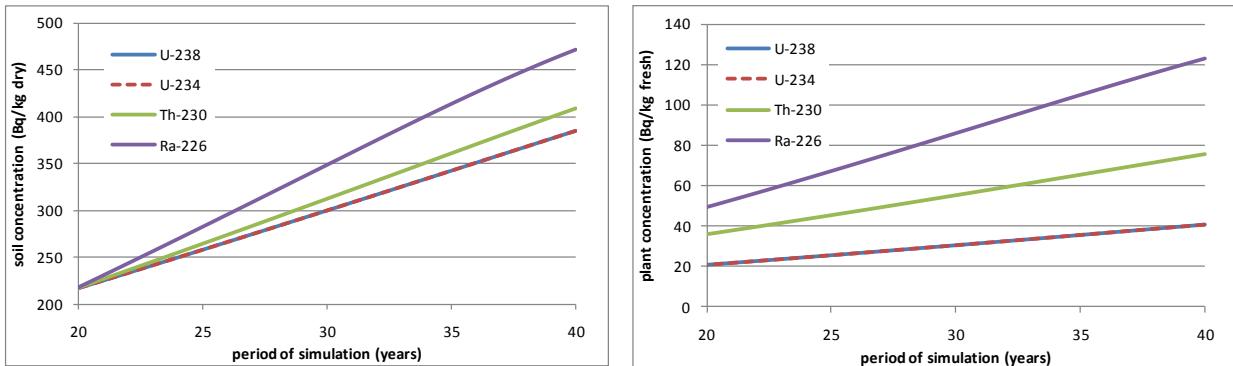


Figure 15 Soil (left) and plant (right) concentrations for the four modelled radionuclides during the 20 year period comparable to 1990 through 2010

8.1.2 HIGH CONCENTRATION SOIL (DOWNSTREAM SITES)

During the first period of simulation from 0 to 19.5 years the required initial contamination of the soil column ranging around $10,000 \text{ Bq kg}^{-1}$ has been reached. The evolution of the calculated soil U-238 concentration through the soil profile is shown in Figure 16. It is considered sufficient to show U-238 since the contamination is identical for the other three radionuclides (Table 23), and because the K_d 's and transfer factors have not changed much for the high concentration soil in comparison to the low concentration case (Table 25).

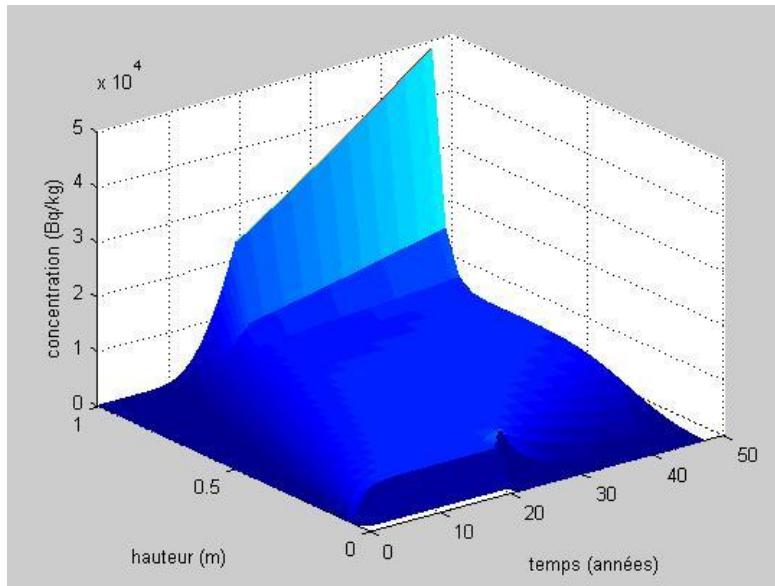


Figure 16 Matlab generated 3-dimension plot depicting the modelled concentration of U-238 in the high concentration soil as a function of time (temps, y) and depth (hauteur, m, with 1 = soil surface). The 27 years of inter-comparison begin with the date 20 years (temps – années).

Because of the high flux of ascending water and the low K_d of $0.0866 \text{ m}^3 \text{ kg}^{-1}$ for U-238 (and the other three radionuclides; see Table 25) a quasi-equilibrium with a maximum at the surface is established in the soil column after this period (Figure 17).

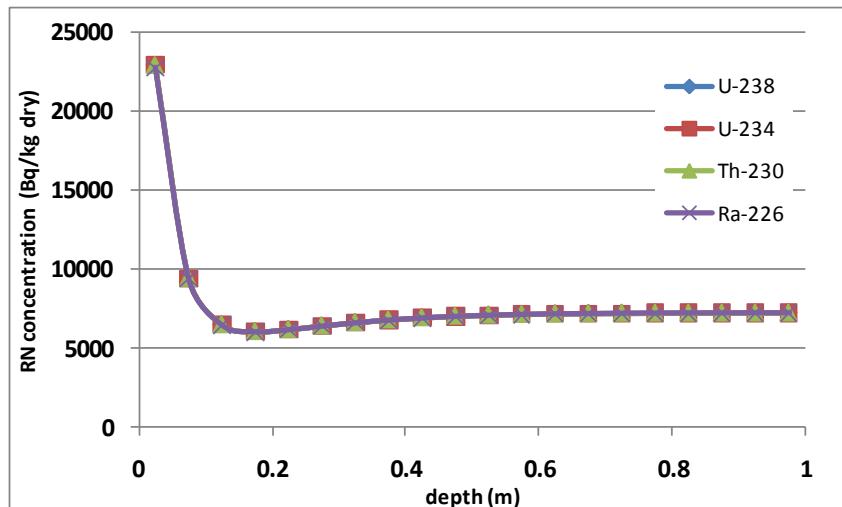


Figure 17 Distribution of the four radionuclides in the soil profile after 19.5 years of initial contamination (0 = surface); giving the onset for the inter-comparison part.

As with the low concentration soil, the concentrations of all four radionuclides are again comparable to what is actually measured in the soil (Table 18). It is from this point on that plant growth is considered and that parameters are derived from the site.

The situation illustrated in the 3-dimensional representation of the soil concentration of the high concentration soil is comparable to the low concentration case. The concentration at depth starts to decrease after 19.5 years to reach "today's" groundwater (river) contamination of 0.3875 Bq L^{-1} (Table 21). The ascending water as well as root uptake and transfer to the surface cause the general upward movement of all radionuclides with time combined with the slow reduction of concentration with depth. As plants are not considered exported, radionuclides taken up by plants are recycled into the first compartment of 0.05 m depth. The concentration with depth at the end of the simulation period shows the significant increase near the surface and the reduction at depth (Figure 18).

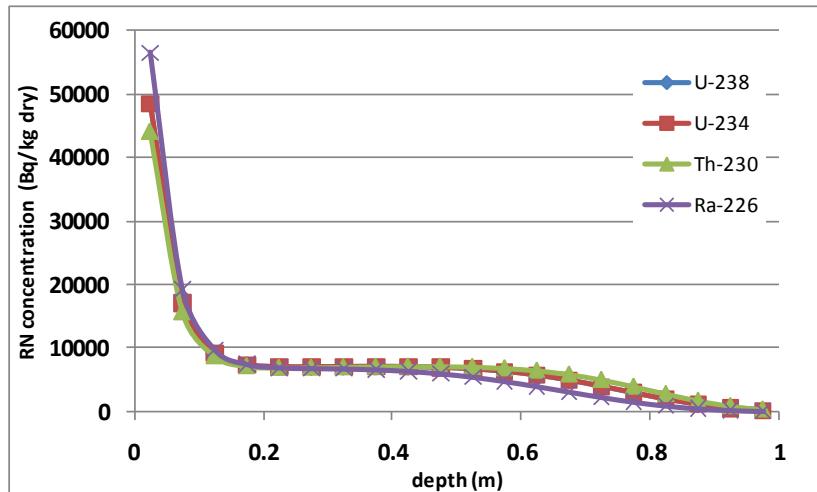


Figure 18 Distribution of the four radionuclides in the soil profile after 47 years of simulation (= 27 years after initial contamination); showing the situation after a period comparable from 1983 to 2010

Finally, Figure 19 presents the results specifically demanded for the inter-comparison exercise, i.e. the concentration of all four radionuclides in the surface layer (0.3 m thickness) and in the plants as a function of time for the period of 1983 to 2010.

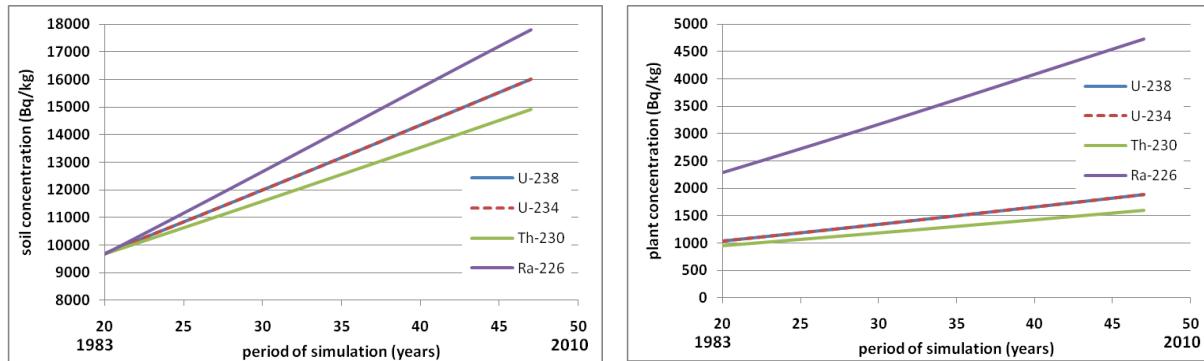


Figure 19 Soil (left) and plant (right) concentration for the four modelled radionuclides during the 27 year period comparable to 1983 through 2010 (high concentration soil case)

8.1.3 CONSIDERATION OF PLANT HARVESTING

ANDRA performed a set of calculations assuming high initial concentrations of radionuclides in the soil, but with 100% loss via plant harvesting (or export), and a much longer simulation period. As shown in Figure 20, such an assumption leads to the calculations indicating a degree of bioremediation occurring at the site.

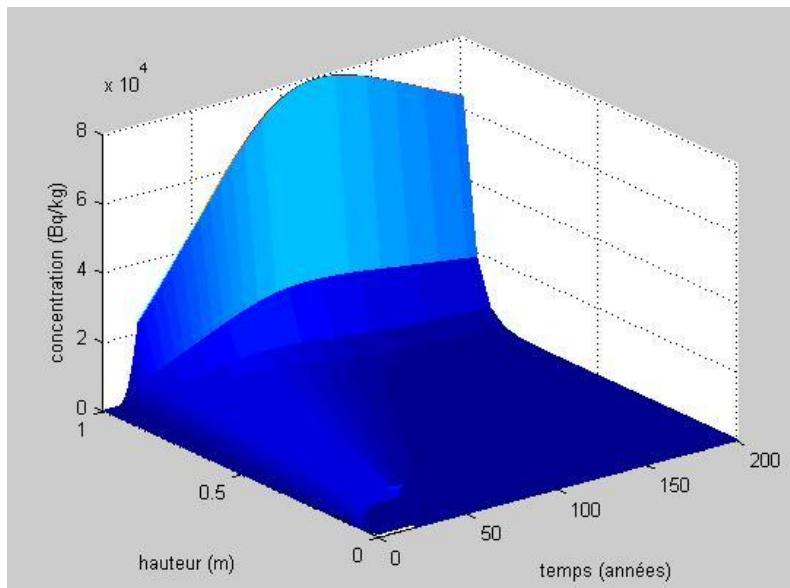


Figure 20 Effect of Plant Harvesting Upon the Long Term Calculated Soil Concentrations of radionuclides

8.2 NDA RWMD BIOSPHERE TOOL

Since the NDA RWMD Biosphere Tool assumes the system is in equilibrium, it was used to calculate soil and plant concentrations that might be associated with the water concentrations measured during the various sampling campaigns (Table 21). The results for the upstream locations (A, B and E) are presented in Table 27, and those for the downstream locations (C and D) in Table 28.

Table 27 Calculated soil and plant radionuclide concentrations in the upstream locations using the NDA RWMD Biosphere Tool (dry weight basis)

| Modelled Variable | Radionuclide | Calculated Result for Given Upstream Location | | |
|--|--------------|---|---------|---------|
| | | A | B | E |
| Surface soil (Bq kg ⁻¹) | U-238 | 3.92E-1 | 1.53E0 | 4.12E-1 |
| | U-234 | 4.65E-1 | 1.81E0 | 4.89E-1 |
| | Th-230 | 1.26E-1 | 8.46E-1 | 1.68E-1 |
| | Ra-226 | 3.85E-1 | 4.24E-1 | 3.81E-1 |
| Sub-soil (Bq kg ⁻¹) | U-238 | 4.26E-1 | 1.66E0 | 4.48E-1 |
| | U-234 | 5.06E-1 | 1.97E0 | 5.31E-1 |
| | Th-230 | 1.37E-1 | 9.20E-1 | 1.83E-1 |
| | Ra-226 | 4.24E-1 | 4.74E-1 | 4.19E-1 |
| Plant (Bq kg ⁻¹) | U-238 | 3.67E-2 | 1.13E-1 | 3.49E-2 |
| | U-234 | 4.36E-2 | 1.33E-1 | 4.14E-2 |
| | Th-230 | 3.69E-2 | 6.33E-2 | 2.73E-2 |
| | Ra-226 | 5.01E-2 | 1.10E-1 | 5.08E-2 |

Table 28 Calculated soil and plant radionuclide concentrations in the downstream locations using the NDA RWMD Biosphere Tool (dry weight basis)

| Modelled Variable | Radio-nuclide | Calculated Result for Given Downstream Location and Sampling Campaign | | | | | | | |
|--|---------------|---|--------|--------|--------|--------|--------|--------|--------|
| | | C | | | | D | | | |
| | | 1 | 2 | 3 | 4 | 1 | 2 | 3 | 4 |
| Surface soil (Bq kg ⁻¹) | U-238 | 3.2E+1 | 6.0E+1 | 2.1E+1 | 1.1E+1 | 3.0E+1 | 5.7E+1 | 2.0E+1 | 1.1E+1 |
| | U-234 | 3.3E+1 | 5.8E+1 | 3.4E+1 | 1.2E+1 | 3.1E+1 | 5.4E+1 | 3.2E+1 | 1.1E+1 |
| | Th-230 | 7.4E-1 | 7.4E-1 | 7.4E-1 | 7.4E-1 | 1.3E0 | 1.3E0 | 1.3E0 | 1.3E0 |
| | Ra-226 | 1.6E+1 | 1.6E+1 | 1.6E+1 | 1.6E+1 | 1.2E+1 | 1.2E+1 | 1.2E+1 | 1.2E+1 |
| Sub-soil (Bq kg ⁻¹) | U-238 | 3.5E+1 | 6.6E+1 | 2.3E+1 | 1.2E+1 | 3.3E+1 | 6.2E+1 | 2.1E+1 | 1.1E+1 |
| | U-234 | 3.6E+1 | 6.3E+1 | 3.7E+1 | 1.3E+1 | 3.4E+1 | 5.9E+1 | 3.4E+1 | 1.2E+1 |
| | Th-230 | 8.0E-1 | 8.0E-1 | 8.0E-1 | 8.0E-1 | 1.4E0 | 1.4E0 | 1.4E0 | 1.4E0 |
| | Ra-226 | 1.8E+1 | 1.8E+1 | 1.8E+1 | 1.8E+1 | 1.4E+1 | 1.4E+1 | 1.4E+1 | 1.4E+1 |
| Plant (Bq kg ⁻¹) | U-238 | 3.3E0 | 6.3E0 | 2.2E0 | 1.2E0 | 3.6E0 | 6.7E0 | 2.3E0 | 1.2E0 |
| | U-234 | 3.4E0 | 6.0E0 | 3.5E0 | 1.3E0 | 3.6E0 | 6.4E0 | 3.7E0 | 1.3E0 |
| | Th-230 | 1.1E-1 | 1.1E-1 | 1.1E-1 | 1.1E-1 | 6.1E-2 | 6.1E-2 | 6.1E-2 | 6.1E-2 |
| | Ra-226 | 2.3E0 | 2.3E0 | 2.3E0 | 2.3E0 | 3.4E0 | 3.4E0 | 3.4E0 | 3.4E0 |

8.3 SYVAC3-CC4

The surface soil and plant concentration in 2010 (27 years after start of simulation) is shown in Table 29 for the five sampling locations (The NWMO model does not calculate concentrations in subsoils).

Table 29 Calculated Concentration in 2010 for Given Location using SYVAC3-CC4 soil model.

| Modelled Variable | Radio-nuclide | Sampling Location | | | | |
|--------------------------------|---------------|-------------------|---------|---------|---------|---------|
| | | A | B | C | D | E |
| Soil Concentration (Bq/kg) | U238 | 9.3E-01 | 4.4E-01 | 1.6E+01 | 1.7E+01 | 1.1E+00 |
| | U234 | 1.0E+00 | 5.0E-01 | 1.8E+01 | 1.9E+01 | 1.1E+00 |
| | Th230 | 9.9E-02 | 1.4E-01 | 1.3E+01 | 6.6E+00 | 1.8E+00 |
| | Ra226 | 1.5E+00 | 6.4E-01 | 4.8E+00 | 1.3E+01 | 1.6E+00 |
| Plant Concentration (Bq/kg) | U238 | 8.7E-02 | 3.2E-02 | 1.2E+00 | 2.0E+00 | 8.9E-02 |
| | U234 | 9.5E-02 | 3.7E-02 | 1.3E+00 | 2.3E+00 | 9.6E-02 |
| | Th230 | 2.9E-02 | 2.1E-02 | 1.0E+00 | 3.1E-01 | 3.0E-01 |
| | Ra226 | 2.0E-01 | 1.7E-01 | 6.8E-01 | 3.5E+00 | 2.2E-01 |

8.4 DISCUSSION OF LOS RATONES RESULTS

The results from the various models are contrasted in this section. It is important to acknowledge that the site specific and source term information has been used and interpreted differently within the different models. In addition, the SAMM model was used to evaluate initial radionuclide levels at the surface, whereas the other models were used only to assess the contamination levels at the surface arising from subsequent migration to the surface. The quantitative comparison of results may be used to help understand the significance of the different data interpretations and ways of representing them, rather than to identify a single best approach.

Focus is given to locations upstream and downstream of the mine, rather than each location individually. The calculated surface soil radionuclide concentrations at the end of the simulation period (2010) are shown in Table 30; the calculated plant radionuclide concentrations for the same time are shown in Table 31. For those models which calculated the soil and plant radionuclide concentration at each of the five locations a mean and range of calculated values is reported.

The SAMM model was used to calculate soil and plant radionuclide concentrations based upon much greater source terms (i.e. groundwater radionuclide concentrations) than those used by the other two models (compare Table 21 and the Table 23 values assumed for SAMM). This reflects different assumptions for the source of the contamination at the surface, i.e. groundwater and river water. Accordingly the results reported from SAMM are based upon groundwater concentrations up to three orders of magnitude greater than those used for the NDA RWMD and SYVAC3-CC4 models.

Once the different groundwater radionuclide concentrations assumed is allowed for, the calculated soil and plant radionuclides concentrations vary within about a factor of three, with the one exception being the NDA RWMD result for Th-230, downstream, which lower than the other results by about an order of magnitude.

Not all routes to explaining the differences have yet been explored. The number of soil layers and those from which the plant takes up any radionuclides varies between these models. Further, there is the choice of differing assumptions about how to treat upward transfer of radionuclides in each model, and as noted earlier in this section the details of the definition of the source term.

Table 30 Comparison of calculated surface soil radionuclide concentrations) at 2010 for the Los Ratones scenario

| Location | Radionuclide | Calculated Soil Radionuclide Concentration at 2010 (Bq kg ⁻¹ dry weight) | | |
|--------------------|--------------|---|----------------------------|---------------------------|
| | | SAMM* | NDA RWMD | SYVAC3-CC4 |
| | | | | |
| Upstream of mine | U-238 | 3.8E+2 | 7.8E-1 (3.9E-1, 1.5E0) | 8.1E-1 (4.4E-1, 1.1E0) |
| | U-234 | 3.8E+2 | 9.2E-1 (4.7E-1, 1.8E0) | 8.8E-1 (5.0E-1, 1.1E0) |
| | Th-230 | 4.1E+2 | 3.8E-1 (1.3E-1, 8.5E-1) | 6.9E-1 (9.9E-2, 1.8E0) |
| | Ra-226 | 4.7E+2 | 4.0E-1 (3.8E-1, 4.2E-1) | 1.3E+0 (6.4E-1, 1.6E0) |
| Downstream of mine | U-238 | 1.6E+4 | 3.0E+1 (1.1E+1, 6.0E+1) | 1.6E+1 (1.6E1, 1.7E1) |
| | U-234 | 1.6E+4 | 3.3E+1 (1.2E+1, 5.8E+1) | 1.9E+1 (1.8E1, 1.9E1) |
| | Th-230 | 1.5E+4 | 1.0E0 (7.4E-1, 1.3E0) | 1.0E+1 (6.6E0, 1.3E1) |
| | Ra-226 | 1.8E+4 | 1.4E+1 (1.2E+1, 1.6E+1) | 8.7E+0 (4.8E0, 1.3E1) |

*Source term assumptions were much higher, see text for explanation.

Table 31 Comparison of calculated plant radionuclide concentrations at 2010 for the Los Ratones scenario

| Location | Radionuclide | Calculated Plant Radionuclide Concentration at 2010 (Bq kg ⁻¹ dry weight) | | |
|--------------------|--------------|--|----------------------------|----------------------------|
| | | SAMM | NDA RWMD | SYVAC3-CC4 |
| | | | | |
| Upstream of mine | U-238 | 4.1E+1 | 6.1E-2 (3.5E-2, 1.1E-1) | 6.9E-2 (3.2E-2, 8.9E-2) |
| | U-234 | 4.1E+1 | 7.3E-2 (4.1E-2, 1.3E-1) | 7.6E-2 (3.7E-2, 9.6E-2) |
| | Th-230 | 7.6E+1 | 4.2E-2 (2.7E-2, 6.3E-2) | 1.2E-1 (2.1E-2, 3.0E-1) |
| | Ra-226 | 1.2E+2 | 7.0E-2 (5.0E-2, 1.1E-1) | 1.9E-1 (1.7E-1, 2.2E-1) |
| Downstream of mine | U-238 | 1.9E+3 | 3.3E0 (1.2E0, 6.7E0) | 1.6E0 (1.2E0, 2.0E0) |
| | U-234 | 1.9E+3 | 3.7E0 (1.3E0, 6.4E0) | 1.8E0 (1.3E0, 2.3E0) |
| | Th-230 | 1.6E+3 | 8.5E-2 (6.1E-2, 1.1E-1) | 6.5E-1 (3.1E-1, 1.0E0) |
| | Ra-226 | 4.7E+3 | 2.9E0 (2.3E0, 3.4E0) | 2.1E0 (6.8E-1, 3.5E0) |

*Source term assumptions were much higher, see text for explanation.

9. SUMMARY AND CONCLUSIONS

This report describes the activities carried out within the project including:

- The development of a generic interaction matrix that details linkages between key features and processes affecting the migration of the radionuclides of interest, U-238, U-234, Th-230 and Ra-226, in surface soils and in crops;
- The description of a range of assessment models;
- The development and modelling of two scenarios, and quantitative estimates of the U-238, U-234, Th-230 and Ra-226 concentrations in specific components of the dose assessment models (soil and plants), for agreed geosphere to biosphere release scenarios of contaminated groundwater, for a hypothetical and an actual site.
- The presentation of results to allow similarities and differences in the different modelling approaches used to evaluate the scenarios.

With respect to the key features and processes affecting the radionuclides considered in this study, they are applicable to a large number of other radionuclides. Those FEPs which were discarded from the IUR FEP matrix in Section 3 generally related to the behaviour of gas, since gases were not of relevance in the current study.

Two types of model have been used in this study:

- conventional radionuclide transfer models with one or two soil compartments, which consider simplistic dynamics of the radionuclides within a soil profile, and
- more complex models that have the potential to account for changes in redox conditions throughout a soil profile (discretised into more than 2 layers), and the effect of redox conditions upon radionuclide behaviour (e.g. sorption).

The models are summarised in Section 4, with the technical details given in Appendix B.

The scenarios, hypothetical and site-specific, were developed specifically for this study. In the case of the hypothetical scenario, use was made of previous model intercomparison studies in the scenario definition (BIOMOVS II, 1995). The set of calculations and results for different precipitation conditions, made using SAMM, illustrate the potential significance of including subsoil layers explicitly within the assessment model, particularly as this affects the representation of upward as well as downward radionuclide movement.

For the site-specific scenario, data was collated from a range of reports and scientific publications produced over the last decade.

Results for the site-specific scenario suggest that the calculated potential radionuclide concentrations in soil and plants vary between models by typically one order of magnitude, and up to a factor of 75. The results also suggest that U-238, U-234, Th-230 and Ra-226 show differences from secular equilibrium of the same order. The models have been developed based on data derived from a range of sites, not just for the Los Ratones site, and so it is not surprising that the range in results is comparable with that observed generally, see UNSCEAR (2006). It is clear that further

characterisation data at depth, and information about the history of activities at the site, would be additionally useful in possible future work.

The more complex models are more data intensive. Further questions which could be considered include: Does that complexity make the results more useful? Does it provide greater insight? When is it necessary to use the more complex approaches? How might site-specific data be used to inform assessment models? For example, future work could consider the implications of being able to estimate or determine site-specific sorption coefficients, based upon information about the soil properties at the site (e.g. Sena et al., 2008; the work by University of Bremen) and upon the estimates of potential impacts of the release of U-238 radionuclides to the biosphere.

Consideration could also be given to the potential importance, or relevance, of accounting for the natural background content of uranium series radionuclides in the soil and water upon the calculated radionuclide concentrations of soil and plants following some form of release as a result of human activities (e.g. mining activities, waste disposal).

There are other avenues of research which the future work of the U-238 series radionuclide working group could consider, dependent upon the wider interests of the group. Consideration could be given to other aspects of the terrestrial ecosystem, such as exposure pathways other than the soil-plant pathway (e.g. inhalation), impacts to non-human biota and doses to humans, or the soil-plant uptake in a non-agricultural ecosystem. If the analysis were extended to comprise freshwater systems, then consideration would need to be given to the process of the accumulation of radionuclides in the freshwater sediments (in rivers and lakes), and the subsequent transfer pathways to aquatic organisms.

This study has focussed upon the upper members of the decay chain. Future work may remain focussed upon the soil-plant system, but considering Ra-226 decay products, notably, Rn-222, Pb-210 and Po-210.

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APPENDIX A: THE IUR GENERAL INTERACTION MATRIX FOR A TERRESTRIAL ECOSYSTEM

The matrix given below is the general interaction matrix developed by the IUR for a terrestrial ecosystem (IUR, 2006). Details of the definitions of contents of the leading diagonal elements and the processes in the off-diagonal elements are provided in this reference.

The letters and numbers used to reference each cell have been added here, and are consistent with the matrices presented in Figure 2 of the main body of this report. The rows and columns associated with “animals” and the “soil atmosphere” feature have been shaded out, and are therefore not numbered, as these features were not considered in the analysis for U, Th and Ra presented in the main body of the report.

BIOPROTA

| | A | B | C | | D | | E | F | G | H |
|---|---|--|---|----------------------------------|--|--|--|-------------------------------|--|--|
| 1 | ATMOSPHERE | 1)Deposition | 1)Deposition 2)Photosynthesis | 1)Inhalation | 1)Dry deposition 2)Precipitation 3)Gas sorption | 1)Diffusive exchange 2)Pressure pumping | | | | 1)Diffusive exchange 2)Pressure pumping (both at outcrop) |
| 2 | 1)Evaporation 2)Gas evolution 3)Droplet production | WATER BODIES | 1)Root uptake 2)Irrigation | 1)Ingestion | 1)Irrigation 2)Recharge by surface waters | 1)Release from solution | | | | 1)Recharge by surface waters |
| 3 | 1)Transpiration 2)Respiration 3)Pollen and seed release 4)Leaf fall 5)Release of other organic matter | 1)Root exudation 2)Senescence and death | VEGETATION | 1)Ingestion | 1)Root exudation | 1)Root respiration | 1)Litter fall 2)Senescence and death | 1)Symbiotic association | | 1)Root exudation 2)Litter fall (at outcrop) 3)Senescence and death 4) Biological weathering |
| | 1)Exhalation 2)Eruption | 1)Excretion 2)Death | 1)Excretion 2)Death | ANIMALS | 1)Excretion | | 1)Excretion 2)Death and decomposition | 1)Excretion of gut microbiota | 1)Excretion | 1)Excretion 2)Death and decomposition (both at outcrop) |
| 4 | 1)Evaporation | 1)Seepage 2)Throughflow 3)Groundwater recharge | 1)Root uptake | 1)Ingestion | SOIL SOLUTION | 1)Ion exchange 2)Degassing | 1)Sorption 2)Fixation | 1)Uptake | 1)Sorption 2)Fixation 3)Diffusion 4)Mineral precipitation | 1)Advection 2)Diffusion |
| | 1)Diffusive exchange 2)Pressure pumping | 1)Solution at boundaries | 1)Root uptake 2) transport in aerenchyma | 1)Inhalation (burrowing animals) | 1)Isotopic exchange 2)Solution | SOIL ATMOSPHERE | 1)Adsorption | 1)Uptake | 1)Adsorption 2)Carbonate production | 1)Diffusive exchange 2)Pressure pumping (both for unsaturated parent material) |
| 5 | 1)Resuspension | | 1)External contamination | 1)Ingestion | 1)Desorption 2)Release during degradation | 1)Degassing | SOIL ORGANIC MATTER | 1)Ingestion 2)Utilisation | 1)Complex formation | 1)Particle transport 2)Colloid transport |
| 6 | 1)Resuspension | | 1)Symbiotic association | 1)Ingestion | 1)Leaching 2)Mineralisation 3)Excretion | 1)Respiration 2)Fermentation | 1)Fertilisation 2)Death and decomposition 3)Biofilms | SOIL MICROBIOTA | | 1)Transport by microbiota 2)Biological weathering |
| 7 | 1)Resuspension | | 1)External contamination | 1)Ingestion | 1)Desorption 2)Mineral dissolution | 1)Degassing | 1)Microbial metabolism | 1)Ingestion 2)Utilisation | SOIL INORGANIC MATTER | 1)Particle transport 2)Colloid transport |
| 8 | 1)Resuspension (at outcrop) | 1)Desorption 2)Mineral dissolution | 1)External contamination | 1)Ingestion 2)Biurbation | 1)Diffusion 2)Capillary rise 3)Colloid transport | 1)Degassing | 1)Microbial metabolism | 1)Ingestion 2)Utilisation | 1)Chemical and mechanical weathering | INTERFACE WITH GEOSPHERE |

APPENDIX B: TECHNICAL DETAILS OF THE MODELS CONSIDERED IN THIS STUDY

Section 4 provides an overview of the models which have been used in this study. The purpose of this appendix is to provide the technical details of these models. All the background information associated with these models is given in the main body of the report.

B.1 CIEMAT REDOX MODEL

The CIEMAT model is fully described in Thorne and Pérez-Sánchez (2010). The model uses a simplified modelling approach to soil hydrology to estimate seasonally varying water contents and fluxes in a multi-layer soil model. The model is implemented in AMBER (Figure 21).

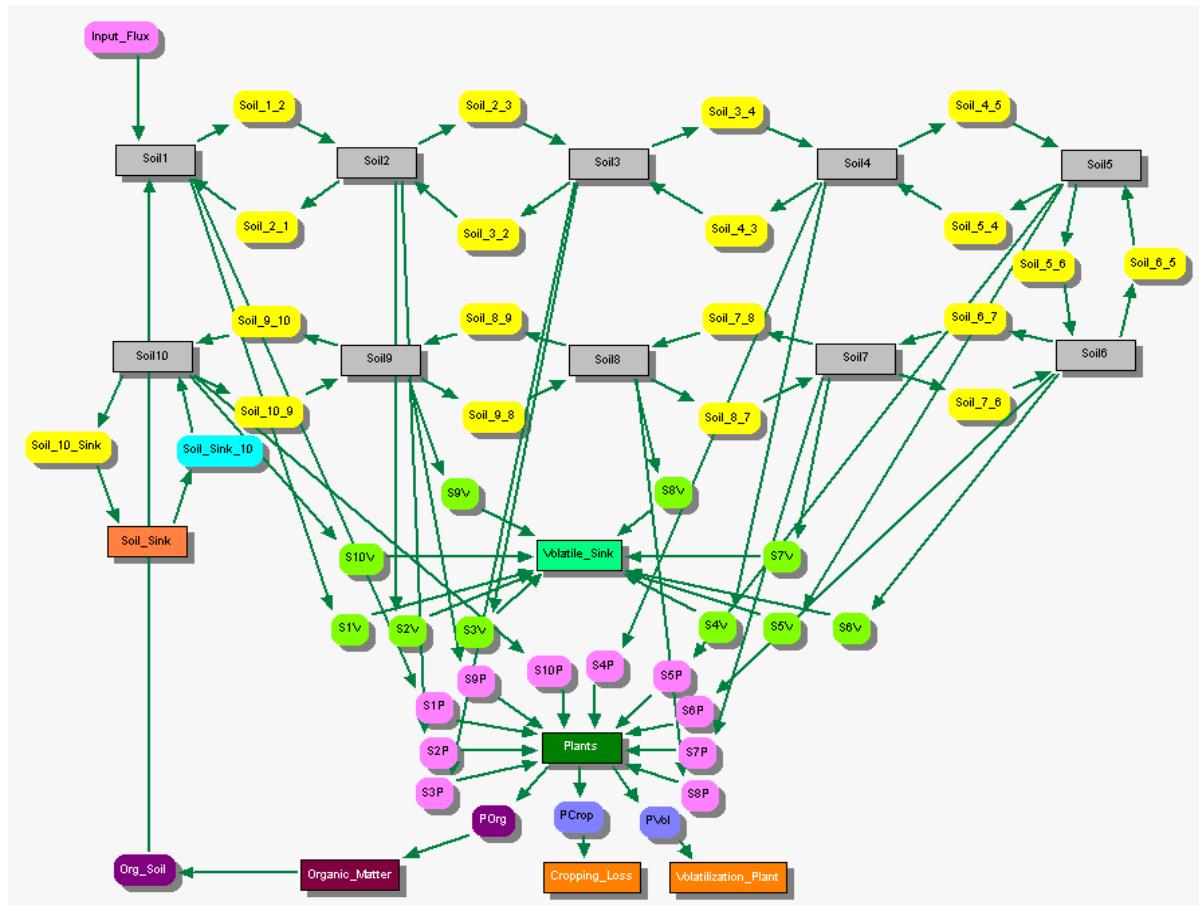


Figure 21 CIEMAT's Soil Redox Model

B.1.1 CALCULATION OF SOIL RADIONUCLIDE CONCENTRATION

For an experiment whose duration is T (y), the concentration of the radionuclide in the soil, C_{soil} (Bq kg^{-1} dw), is given by the following equation.

$$C_{\text{soil}} = C_{\text{soil},0} \cdot \exp(-\lambda_p \cdot T) \quad (1)$$

Here $C_{\text{soil},0}$ is the radionuclide concentration in the soil at the start of the experiment, and λ_p is the transfer rate from the soil to the plant given by:

$$\lambda_p = \rho_r \cdot d \cdot A \cdot f(\theta) \cdot K \quad (2)$$

Here:

- ρ_r is the length of fine roots in the soil (m root per m^3 soil);
- d is the depth of the soil (m);
- A is the model area (1 m^2);
- $f(\theta)$ is a function of varying the effectiveness of root uptake with the water content of the soil (-);
- K is a normalisation coefficient defined as the fractional uptake rate per unit length of fine roots ($\text{m}^{-1} \text{ y}^{-1}$).

B.1.2 CALCULATION OF PLANT RADIONUCLIDE CONCENTRATION

Plants are assumed to take up radionuclides from their roots only. Radionuclides are lost from the plants via cropping. In addition, as the model was originally developed for Se-79, the model also considers volatilisation losses from plants. If the standing biomass density at the end of the experiment is Y (kg dw m^{-2}) and the duration of the experiment is T (y), then the concentration in plants at that time, C_{plant} (Bq kg^{-1} dw) is given by:

$$C_{\text{plant}} = A \cdot C_{\text{soil},0} \cdot \rho_s \cdot d \cdot \frac{[1 - \exp(-\lambda_p \cdot T)]}{A \cdot Y} \quad (3)$$

Here ρ_s is the dry bulk density of the soil (kg m^{-3}), and the other parameters are as defined above.

B.2 ANDRA'S SAMM MODEL

The model SAMM allows simulation of transfers from a source within the geosphere (defined by full water saturation) to the soil surface and the plant via a variety of physical transport phenomena, such as diffusion, advection and root transfer.

B.2.1 MODEL STRATEGY

The model uses a discretisation in time and space with the latter based on a 1D approach using a defined number of compartments (Figure 22).

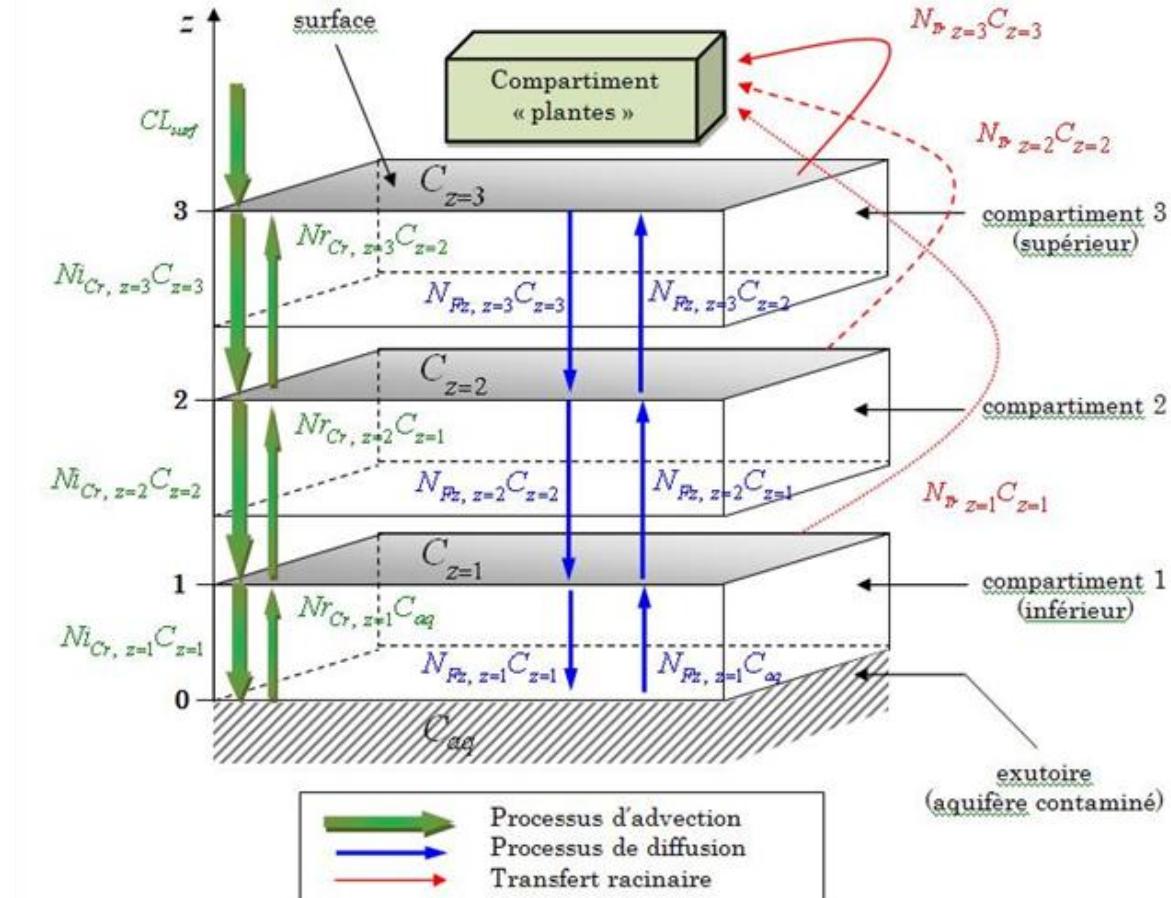


Figure 22 Representation of a 3 compartment setup illustrating discretisation of the model in space between the geosphere (i.e. an aquifer - C_{aq}) and the surface soil layer with its associated plant compartment (Compartiment "plantes"). Transfer can be advective up or down (green arrows), diffusive (blue arrows) or via root transport to the plant and the soil surface (red arrows) and are governed by the Fourier and the Courant numbers (N_F , Nr_{CR}).

B.2.1 MODEL EQUATIONS

The classical advection diffusion equation forms the basis for the mathematical treatment of the transport phenomena:

$$\begin{aligned}
\frac{\partial C_{V_eau}(z,t)}{\partial t} = & \underbrace{Vi_z(z,t) \frac{\partial C_{V_eau}(z,t)}{\partial z} + C_{V_eau}(z,t) \frac{\partial Vi_z(z,t)}{\partial z}}_{\text{downward advection}} \\
& \underbrace{- Vr_z(z,t) \frac{\partial C_{V_eau}(z,t)}{\partial z} + C_{V_eau}(z,t) \frac{\partial Vr_z(z,t)}{\partial z}}_{\text{upward advection}} \\
& \underbrace{+ D_a(z) \frac{\partial^2 C_{V_eau}(z,t)}{\partial z^2} + \frac{\partial D_a(z)}{\partial z} \frac{\partial C_{V_eau}(z,t)}{\partial z}}_{\text{diffusion}} \\
& \underbrace{- \lambda \cdot C_{V_eau}(z,t) + \lambda_p \cdot \frac{[\theta + \rho_b \cdot Kd_{sol_p}]}{[\theta + \rho_b \cdot Kd_{sol}]} C_{V_eau_p}(z,t)}_{\text{radioactive decay}} \\
& \underbrace{- d_r(z) \cdot r_v \cdot FT_{sp} \left(\frac{\theta}{\rho_b} + Kd_{sol} \right) C_{V_eau}(z,t)}_{\text{root uptake}}
\end{aligned} \tag{4}$$

It is completed using an equation for radioactive decay and in growth and root / plant uptake. Downward advection occurs if the hydraulic balance of the soil (precipitation plus irrigation minus evapotranspiration) is positive. Upward advection (i.e. via capillary rise) occurs if evapotranspiration is greater than input. Diffusion is controlled by the local concentration gradients of the considered substances (radionuclide or toxic chemical).

The transfer of radionuclides and toxic chemicals to plants is a function of the soil – plant transfer factor (I), the root depth distribution (II) and the total soil concentration (III):

$$C_{M_pl}(t) = \int_0^z d_r(z) \cdot FT_{sp} \left(\underbrace{\frac{\theta}{\rho_b} + Kd_{sol}}_{III} \right) C_{V_eau}(z,t) dz \tag{5}$$

Detailed information on the parameters used in the equation are summarised in Table 30.

Table 30 SAMM Model Parameters

| Parameter | Description | Dimension |
|----------------------|--|---|
| $C_{V_eau}(z,t)$ | Concentration of radionuclide or toxic chemical in the soil water a function of space and time | Bq m ⁻³ |
| $Vi_z(z,t)$ | Velocity of advective downward flow as a function of space and time | m y ⁻¹ |
| $Vr_z(z,t)$ | Velocity of advective upward flow as a function of space and time | m y ⁻¹ |
| $D_a(z,t)$ | Apparent diffusion coefficient as a function of position and time | |
| λ | Disintegration constant for the radionuclide (daughter) | y ⁻¹ |
| $C_{V_eau_p}(z,t)$ | Concentration of the parent radionuclide in the soil water | Bq m ⁻³ |
| λ_p | Disintegration constant for the parent radionuclide | y ⁻¹ |
| θ | Soil water content | (-) |
| ρ_b | Soil density | kg m ⁻³ |
| Kd_{sol} | Solid – solution distribution coefficient for the radionuclide or toxic chemical | m ³ kg ⁻¹ |
| Kd_{sol_p} | Solid – solution distribution coefficient for the parent radionuclide | m ³ kg ⁻¹ |
| $d_r(z)$ | Root distribution as a function of depth z | (-) |
| r_V | Volume-related yield of plants | kg (fw) m ⁻³ y ⁻¹ |
| FT_{sp} | Soil – plant transfer factor | kg (dw) kg ⁻¹ (fw) |

The differential equations are solved in Matlab using numeric discretisation (Taylor-Young formulation and finite differences); input data are read directly from an Excel sheet. Output data can either be evaluated further within Matlab or exported to an Excel output sheet.

B.3 NDA RWMD BIOSPHERE ASSESSMENT TOOL

The key equations for the equilibrium case are listed below. The water balance in the soil system is described by:

$$I_1 + E_2 - I_2 - E_1 - S_{out} = 0 \quad (6)$$

$$G + I_2 - E_2 - B_{out} = 0 \quad (7)$$

Radionuclide transport in the soil system is described by:

$$E_2 \left(\frac{C_2}{K_2} \right) - \left(S_{out} + I_2 \right) \left(\frac{C_1}{K_1} \right) - \lambda d_I C_I + \lambda d_I C_{I,parent} + F_I = 0 \quad (8)$$

$$I_2 \left(\frac{C_1}{K_1} \right) - (E_2 + B_{out}) \left(\frac{C_2}{K_2} \right) - \lambda d_2 C_2 + \lambda d_2 C_{2,parent} + F_2 = 0 \quad (9)$$

$$K_1 = \phi_1 + \rho_1 K_{d1} \quad (10)$$

$$K_2 = \phi_2 + \rho_2 K_{d2} \quad (11)$$

In the kinetic approach implemented in GoldSim, the first two of these transport equations are substituted by their kinetic form in which the time derivatives of the concentrations C_1 and C_2 replace the zeros on the right hand side.

In these equations, the variables are as defined in Table 31. The hydrological variables are shown explicitly in Figure 23. Typically in application, the interface with the underlying geosphere model will be at rockhead, i.e. the total depth of the surface soil plus subsoil will be taken equal to the depth of unconsolidated material. However, this is not a requirement of the model and the total depth of surface soil plus subsoil may be less than the depth of unconsolidated material.

Table 31 Definitions of Model Variables

| Quantity | Units | Description |
|----------------|----------------------------------|---|
| I_1 | m y^{-1} | Precipitation plus irrigation |
| I_2 | m y^{-1} | Percolation to subsoil |
| E_1 | m y^{-1} | Evapotranspiration |
| E_2 | m y^{-1} | Capillary rise to replenish soil moisture deficit |
| S_{in} | m y^{-1} | Throughflow gains from surface soil |
| B_{in} | m y^{-1} | Baseflow gains from subsoil |
| S_{out} | m y^{-1} | Throughflow losses from surface soil |
| B_{out} | m y^{-1} | Baseflow losses from subsoil |
| G | m y^{-1} | Groundwater discharge to subsoil |
| C_1 | Bq m^{-3} | Concentration of the radionuclide of interest in surface soil |
| C_2 | Bq m^{-3} | Concentration of the radionuclide of interest in subsoil |
| λ | y^{-1} | Decay constant of the radionuclide of interest |
| d_1 | m | Depth of the surface soil layer |
| d_2 | m | Depth of the subsoil layer |
| $C_{1,parent}$ | Bq m^{-3} | Concentration of the immediate parent of the radionuclide of interest in surface soil |
| $C_{2,parent}$ | Bq m^{-3} | Concentration of the immediate parent of the radionuclide of interest in subsoil |
| F_1 | $\text{Bq m}^{-2} \text{y}^{-1}$ | Flux of the radionuclide of interest into surface soil |
| F_2 | $\text{Bq m}^{-2} \text{y}^{-1}$ | Flux of the radionuclide of interest into subsoil |
| ϕ_1 | - | Water-filled porosity of surface soil |
| ρ_1 | kg m^{-3} | Dry bulk density of surface soil |
| K_{d1} | $\text{m}^3 \text{kg}^{-1}$ | Distribution coefficient for the radionuclide of interest in surface soil |
| ϕ_2 | - | Water-filled porosity of subsoil |
| ρ_2 | kg m^{-3} | Dry bulk density of subsoil |
| K_{d2} | $\text{m}^3 \text{kg}^{-1}$ | Distribution coefficient for the radionuclide of interest in subsoil |

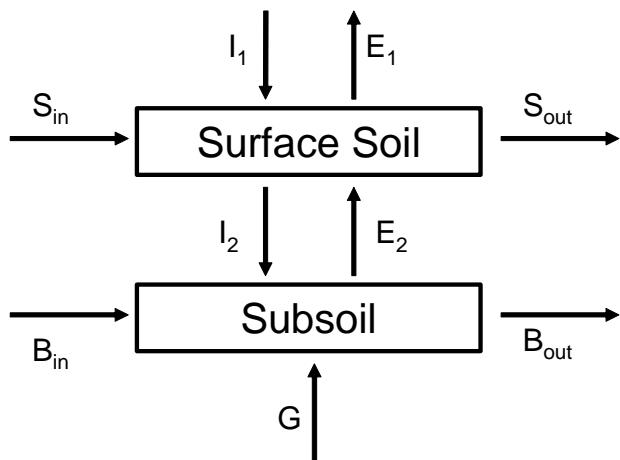


Figure 23 Hydrological Quantities used in the Soil Model

For natural groundwater discharge, $F_1 = 0$ and F_2 is taken as a constant (often 1.0, but this is not required in the spreadsheet implementation). For well abstraction and subsequent irrigation, $F_2 = 0$ and $F_1 = I_{\text{irr}} * C_w$, where I_{irr} (m y^{-1}) is the irrigation rate and C_w (Bq m^{-3}) is the concentration of the radionuclide of interest in the abstracted well water (often taken as 1.0, but this is not required in the spreadsheet implementation).

In evaluating the radionuclide transport equations, it is convenient to define the following supplementary quantities:

$$T_1 = F_1 + \lambda d_1 C_{1,\text{parent}} \quad (12)$$

$$T_2 = F_2 + \lambda d_2 C_{2,\text{parent}} \quad (13)$$

$$\alpha_1 = \frac{E_2}{K_2} \quad (14)$$

$$\alpha_2 = \frac{E_2 + B_{\text{out}}}{K_2} + \lambda d_2 \quad (15)$$

$$\beta_1 = \frac{S_{\text{out}} + I_2}{K_1} + \lambda d_1 \quad (16)$$

$$\beta_2 = \frac{I_2}{K_1} \quad (17)$$

With these definitions, the transport equations become:

$$\beta_1 C_1 - \alpha_1 C_2 = T_1 \quad (18)$$

$$-\beta_2 C_1 + \alpha_2 C_2 = T_2 \quad (19)$$

B.3.1 CALCULATION OF SOIL RADIONUCLIDE CONCENTRATION

Consideration of the transport equations given above allows the total concentrations of the radionuclide to be determined:

$$C_1 = \frac{\alpha_1 T_2 + \alpha_2 T_1}{\alpha_2 \beta_1 - \alpha_1 \beta_2} \quad (20)$$

$$C_2 = \frac{\beta_1 T_2 + \beta_2 T_1}{\alpha_2 \beta_1 - \alpha_1 \beta_2} \quad (21)$$

C_1 and C_2 are the total concentrations in soil, expressed on a volumetric basis. They are converted to a dry mass basis (Bq kg^{-1}) using:

$$CT_1 = \frac{C_1}{\rho_1} \quad (22)$$

$$CT_2 = \frac{C_2}{\rho_2} \quad (23)$$

Also, the concentrations on soil solids and in soil solution are required separately. Concentrations in soil solids (Bq kg^{-1}) are given by:

$$CS_1 = \frac{CT_1 \rho_1 K_{d1}}{\phi_1 + \rho_1 K_{d1}} \quad (24)$$

$$CS_2 = \frac{CT_2 \rho_2 K_{d2}}{\phi_2 + \rho_2 K_{d2}} \quad (25)$$

Concentrations in soil solution (Bq m^{-3}) are given by:

$$CL_1 = \frac{C_1}{\phi_1 + \rho_1 K_{d1}} \quad (26)$$

$$CL_2 = \frac{C_2}{\phi_2 + \rho_2 K_{d2}} \quad (27)$$

In undertaking the calculations for decay chains, the calculations are performed in sequence for the radionuclides. Thus, the first calculation is for the radionuclide at the head of the chain, for which contributions from a predecessor radionuclide are set to zero. Values of C_1 and C_2 for this radionuclide are the concentrations of parent required for the next member of the chain. This process

is then iterated down the whole chain. Note that only the longer-lived members of chains are computed explicitly. Shorter-lived, intermediate radionuclides are assumed to be present in secular equilibrium and the various external and internal dose factors used take their contributions into account. Note also that F_1 and F_2 are set to zero for all members of a chain except the first. This ensures that the results obtained relate to discharge of the first member of the chain alone, but include contributions from the ingrowth of its progeny within the biosphere system.

B.3.2 CALCULATION OF PLANT RADIONUCLIDE CONCENTRATION

The contamination of plants via root uptake and irrigation are explicitly considered in the model. The deposition of contaminated dust particles on the plant is considered implicitly in the model via the consumption of soil by both animals and humans, but it does not contribute directly to the contamination of plants.

The radionuclide concentration in the plant as a result of root uptake is calculated by multiplying the radionuclide concentration in soil by a concentration ratio, $T_{n,r}$ (kg (dw) soil per kg (fw) vegetation).

$$C_{plant,root,r} = C_{soil,r} \cdot T_{n,r} \quad (28)$$

The approach to calculating the radionuclide concentration in the plant as a result of the interception of irrigation water is

$$C_{plant,intercept} = F_{intercept} \cdot \frac{C_{water} \cdot V_{irr}}{Y} \cdot [F_{ext} \cdot [1 - F_{abs}] \cdot \exp(-k \cdot T) + F_{abs} \cdot F_{int} \cdot F_{transloc}] \quad (29)$$

Here:

- $F_{intercept}$ is the irrigation water interception factor (-);
- C_{water} is the concentration of the radionuclide in the irrigation water (Bq m^{-3});
- V_{irr} is the annual irrigation rate (m y^{-1});
- Y is the yield of the edible component of the vegetation ($\text{kg (fw) m}^{-2} \text{y}^{-1}$);
- F_{abs} is the fraction of the intercepted activity that is absorbed by the vegetation (-);
- $F_{transloc}$ is the fraction of the absorbed activity that is translocated to the edible part (-);
- F_{int} is the fraction of absorbed activity translocated to the edible part that is retained after food processing (-);
- F_{ext} is the fraction of external contamination that remains associated with the edible part after food processing (-);
- k is the wash-off coefficient (d^{-1}); and
- T is the number of days between irrigation and harvest (d).

The interception factor, $F_{intercept}$, is calculated using the Chamberlain formula (Chamberlain, 1970):

$$F_{\text{intercept}} = 1 - e^{-\mu w} \quad (30)$$

where μ ($\text{m}^2 \text{ kg}^{-1}$ dry weight) is known as the interception coefficient and w (kg (dry weight) m^{-2}) is the standing above-ground biomass density at the time of irrigation. Combining the above expressions, for plants contaminated both by root uptake and spray irrigation, the appropriate form is:

$$C_{\text{plant}} = \eta_{\text{plant}} C_{\text{soil}} + [1 - \exp(-\mu w)] \left(I \frac{C_w}{Y} \right) \times [F_{p3}(1 - F_{\text{abs}}) \exp(-kT) + F_{\text{abs}} F_{p2} F_{\text{trans}}] \quad (31)$$

B.4 SYVAC3-CC4

The transport processes considered in the soil sub-model of SYVAC3-CC4 are shown in Figure 24.

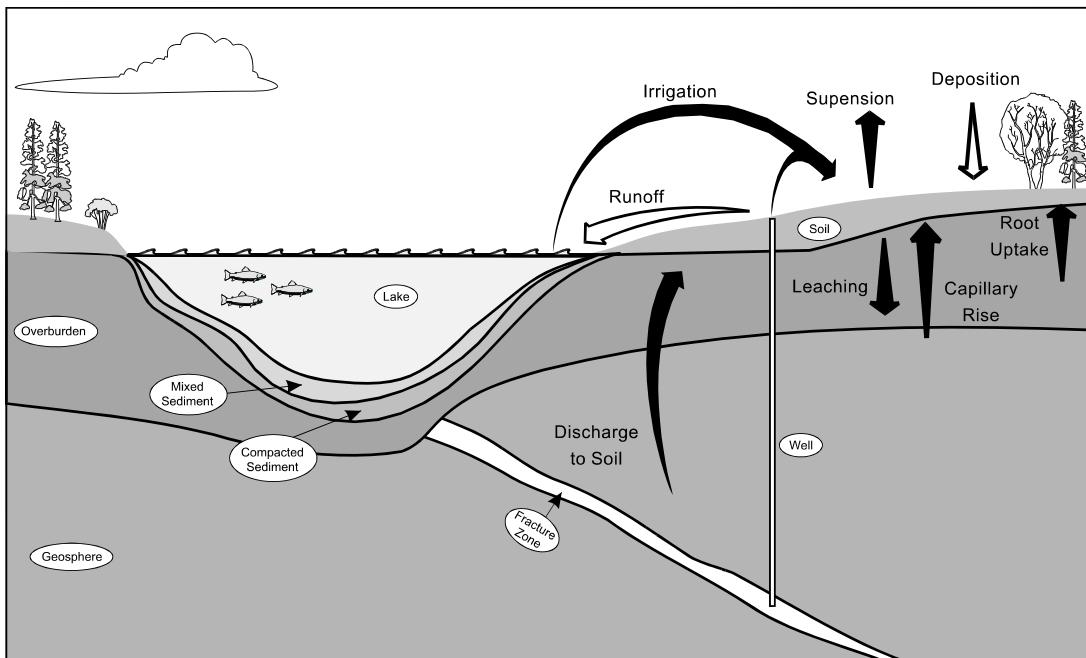


Figure 24: Transport Processes Considered in the SYVAC3-CC4 Soil Sub-model.

In the adaptation of the model for the Los Ratones scenario description, there was no well and the runoff was ignored.

The basic surface soil contaminant model is:

$$\frac{dA_s^{i,f}(t)}{dt} = X_d^{i,f}(t) + X_{bs}^{i,f}(t) + X_{irr}^{i,f}(t) + \lambda^p A_s^{p,f}(t) - (\lambda_{le}^{i,f} + \lambda^i) \cdot A_s^{i,f}(t) \quad (32)$$

Where:

$A_s^{i,f}(t)$ is the amount of nuclide i in the surface soil compartment for sampling point f at time t (mol)

$X_d^{i,f}(t)$ is the nuclide i input flow from deposition from the atmosphere for sampling point f at time t (mol y^{-1})

$X_{bs}^{i,f}(t)$ is the input from upward movement of nuclide i with groundwater into the surface soil for sample point f at time t (mol y^{-1})

$X_{irr}^{i,f}(t)$ is the nuclide i input flow from irrigation to the sampling point f at time t (mol y^{-1})

λ^p is the decay rate of any parent radionuclide p (y^{-1})

$A_s^{p,f}(t)$ is amount of parent nuclide p in the soil at time t for sampling point f (mol)

$\lambda_{le}^{i,f}$ is the nuclide loss rate due to leaching at sampling point f (y^{-1})

λ^i is the loss due to radioactive decay of nuclide i (y^{-1})

Soil erosion, bioturbation, and volatilization are not considered as removal terms in the CC4 model.

B.4.1 CALCULATION OF SOIL RADIONUCLIDE CONCENTRATION

The soil model calculates the concentration of contaminants in the surface (rooting or cultivated) soil layer, which is assumed to be well-mixed (e.g. plowing, bioturbation). Soil can become contaminated as a result of atmospheric deposition, capillary rise or the application of irrigation water to the field.

The input from deposition from the atmosphere (mol y^{-1}) is calculated with the following equations:

$$X_d^{i,f}(t) = D_{soil}^{i,f}(t) A_F \quad (33)$$

$$D_{soil}^{i,f}(t) = (C_{air,AG}^i + C_{air,AP}^{i,f}) \cdot (V_d + P_{tot} \cdot W_r) \quad (34)$$

$$C_{air,AP}^i = A_{DLA} \cdot C_L^{i,f} \quad (35)$$

$$C_{air,AG}^i \propto \lambda_{vol}^i \quad (36)$$

Where:

$X_d^{i,f}(t)$ is the input from nuclide i deposition from the atmosphere at time t (mol y^{-1})

$D_{soil}^{i,f}(t)$ is the deposition rate per unit area of nuclide i to the soil at time t for sampling point f (mol $m^{-2} y^{-1}$)

A_F is the area of the field (m^2)

$C_{air,AG}^i$ is the outdoor air concentration of nuclide i from aquatic degassing or volatilization ($\text{mol m}^{-3}_{\text{air}}$)

$C_{air,AP}^{i,f}$ is the outdoor air concentration of nuclide i from aquatic particulate or spray sources for sampling point f ($\text{mol m}^{-3}_{\text{air}}$)

V_d is the dry deposition velocity (m y^{-1})

P_{tot} is the total annual precipitation rate (m y^{-1})

W_r is the atmosphere washout ratio (-)

A_{DLA} is the atmospheric particle dust load from aquatic sources ($\text{m}^3_{\text{water}} \text{m}^{-3}_{\text{air}}$)

$C_L^{i,f}$ is the concentration of nuclide i in the surface water near sampling point f .

Because the water-to-air volatilization loss rate constant λ_{vol}^i is zero for all nuclides considered here, the outdoor air concentration from aquatic sources $C_{air,AG}^i$ is also zero.

The subsoil to surface soil nuclide transport is described by capillary rise from the contaminated groundwater. This is the dominant input pathway at the Los Ratones site, for surface soils that are not initially surface contaminated. Nuclide flow due to capillary rise is described by the following equations:

$$X_{bs}^{i,f} = C_{gw}^{i,f} Q_{bs} A_F \quad (37)$$

$$Q_{bs} = Q_{swd} \left(\frac{Z_{ss}}{Z_d} \right)^b, \text{ if } Z_d \geq Z_{ss} \quad (38)$$

Where:

$X_{bs}^{i,f}$ is the nuclide i flow to the sample point (mol y^{-1})

$C_{gw}^{i,f}$ is the nuclide i groundwater pore water concentration at sampling point f ($\text{mol m}^{-3}_{\text{water}}$)

Q_{bs} is the capillary water flow from the water table through the subsoil to the surface soil ($\text{m}^3 \text{y}^{-1}$)

Q_{swd} is the summer water deficit [ET-P] ($\text{m}^3_{\text{water}} \text{m}^{-2}_{\text{soil}} \text{y}^{-1}$)

Z_d is the depth to the water table (m)

The river water is assumed to be the source of irrigation. Nuclide flow due to irrigation is described by the following equation.

$$X_{irr}^{i,f}(t) = I_{irr}^f \cdot A_F \cdot C_{irr}^{i,f}(t) \quad (39)$$

Where:

$X_{irr}^{i,f}(t)$ is the nuclide i input flow from irrigation to the sample soil field f at time t (mol y^{-1})

I_{irr}^f is the rate of irrigation for each sampling point f ($\text{m}^3_{\text{water}} \text{ m}^{-2}_{\text{soil}} \text{ y}^{-1}$)

$C_{irr}^{i,f}(t)$ is the nuclide i concentration of the irrigation water to each sampling point f at time t (mol m^{-3})

Radionuclides are assumed to be lost from the soil-plant system via leaching.

Loss by leaching deals with the downward migration through the soil in solution on colloidal particles or dissolved in infiltrating water. The loss due to leaching at sample point f [a^{-1}] is expressed as follows:

$$\lambda_{le}^{i,f} = \frac{f_{le} (Q_{bs} + I_{irr}^f + R_T)}{(\theta_s^f + \rho_{soil}^f \cdot K_d^{i,f}) \cdot Z_{ss}} \quad (40)$$

f_{le} is the leaching rate fraction

R_T is the average watershed runoff

θ_s^f is the volumetric water content of the surface soil for sampling point f .

ρ_{soil}^f is the dry soil bulk density for sampling point f .

$K_d^{i,f}$ is the sorption coefficient of nuclide i in each sampling point f .

Z_{ss} is the depth of the surface soil layer (m)

The resulting nuclide concentration in the surface soil compartment is then expressed as:

$$C_{soil}^{i,f} = \frac{A_s^{i,f}(t)}{A_F Z_{ss}} \quad (41)$$

B.4.2 CALCULATION OF PLANT RADIONUCLIDE CONCENTRATION.

Plants are assumed to become contaminated via root uptake. The plant radionuclide concentration is therefore calculated using the following expression:

$$C_{plant}^{i,f} = C_{soil} \cdot CR^{i,f} \quad (42)$$

$CR^{i,f}$ is the plant/soil concentration ratio for nuclide i and sampling point f .

B.5 RESRAD-OFFSITE

CALCULATION OF PLANT RADIONUCLIDE CONCENTRATION

Plants are assumed to become contaminated via root uptake (from either onsite or offsite soil), foliar uptake from overhead irrigation, and foliar uptake of dust.

Root uptake: If part (or all) of the agricultural area lies above the primary contamination, the roots of the plants could penetrate the primary contamination and take up the contaminants. This is computed as follows:

$$p_i^{onsite}(t) = f_a \cdot f_{cd}(t) \cdot rtf_i \cdot s_i(t) \quad (43)$$

Here:

- $p_i^{onsite}(t)$ is the activity concentration of radionuclide i in the edible portion of the plant resulting from root uptake from the area of primary contamination (pCi g^{-1});
- f_a is the fraction of the agricultural area that lies directly above the primary contamination (-);
- rtf_i is the root uptake factor of radionuclide i for the plant (-);
- $s_i(t)$ is the activity concentration of radionuclide i in onsite soil (pCi g^{-1}); and
- $f_{cd}(t)$ is the cover and depth factor, which is the fraction of the root length exposed to the primary contamination (-).

$f_{cd}(t)$ is calculated as follows:

$$\begin{aligned} f_{cd}(t) &= 0 && \text{when } T_{cv}^c(t) \geq d_{root} \\ &= f_{vm}(t) \frac{\rho_{pc}}{\rho_{mix}} && \text{when } T_{mix}^c(t) \geq d_{root} \\ &= \frac{f_{vm}(t) \frac{\rho_{pc}}{\rho_{mix}} T_{mix}^c(t) + d_{root} - T_{cv}^c(t) - T_{cz}^{um}(t)}{d_{root}} && \text{when } T_{cv}^c(t) + T_{mix}^c(t) + T_{pc}^{um}(t) \geq d_{root} \quad (44) \\ &= \frac{f_{vm}(t) \frac{\rho_{pc}}{\rho_{mix}} T_{mix}^c(t) + T_{cz}^{um}(t)}{d_{root}} && \text{when } T_{cv}^c(t) + T_{mix}^c(t) + T_{pc}^{um}(t) < d_{root} \end{aligned}$$

Here:

- f_{vm} is the volume fraction of primary contamination in the mixing zone (-);
- ρ_{pc} is the dry bulk density of the primary contamination (g cm^{-3});
- ρ_{mix} is the dry bulk density of the mixing zone (g cm^{-3});
- $T_{mix}^c(t)$ is the thickness of the contaminated mixing layer (m);
- $T_{cv}^c(t)$ is the thickness of the clean cover (m);
- $T_{pc}^{um}(t)$ is the thickness of the unmixed portion of the primary contamination (m); and
- d_{root} is the length of the root (m).

The volume fraction of primary contamination in the mixing zone, f_{vm} , is a dynamic parameter; its evolution with time is described in Yu et al. (2007).

The activity concentration of radionuclide i in the edible portion of the plant result from uptake by roots from the offsite soil is computed as follows:

$$p_i^{offsite}(t) = r f_i \cdot s_i^o(t) \quad (45)$$

Adjustments are not made for the situation where the depth of root might exceed the thickness of the mixing layer because the code only models the concentration of radionuclides in the mixing zone, not in the soil below.

Foliar uptake from overhead irrigation: This uptake is calculated as follows:

$$p_i^{oi}(t) = \frac{f_{int} \cdot f_{tl}}{Y} \cdot \frac{1 - \exp(-\lambda_w t_g)}{\lambda_w} \cdot \frac{q_{ir}}{t_g} \cdot w_i^{ir}(t) \quad (46)$$

Here $p_i^{oi}(t)$ is the activity concentration of radionuclide i in the edible portion of the plant resulting from foliar interception of overhead irrigation (pCi g^{-1}), f_{tl} is the fraction of contaminant that is translocated from the foliage to the edible parts of the plants, and Y is the wet weight crop yield (kg m^{-2}).

This expression is derived by assuming that the concentration of radionuclide in the irrigation water is constant during the growing season. Consider the changes in the quantity of the radionuclide in the plant over a time period of δt . The quantity of the radionuclide intercepted and retained by the plant is given by

$$f_{int} \cdot \frac{q_{ir}}{t_g} \cdot w_i^{ir} \delta t \quad (47)$$

The quantity of the radionuclide lost from the plant due to weathering is given by:

$$\lambda_w \cdot P_i^{oi}(t) \cdot \delta t \quad (48)$$

Here $P_i^{oi}(t)$ is the quantity of radionuclide in the plant grown in a unit area of land, resulting from overhead irrigation. The change in the quantity of the radionuclide in the plant is represented by:

$$\frac{P_i^{oi}(t)}{dt} \cdot \delta t \quad (49)$$

Equating the change to the net change gives the mass balance equation:

$$\frac{P_i^{oi}(t)}{dt} = f_{int} \cdot \frac{q_{ir}}{t_g} \cdot w_i^{ir} - \lambda_w \cdot P_i^{oi}(t) \quad (50)$$

Integration over the growing period gives:

$$P_i^{oi}(t) = f_{int} \cdot \frac{q_{ir}}{t_g} \cdot w_i^{ir} \cdot \frac{1 - \exp(-\lambda_w t_g)}{\lambda_w} \quad (51)$$

This then accounts for the quantity of the radionuclide that is retained by the plant. The concentration in the edible portion of the plant is obtained by applying the translocation factor and yield.

Foliar uptake of dust: This uptake is computed as follows:

$$p_i^{dust}(t) = \frac{f_{int} \cdot f_{tl}}{Y} \cdot \frac{1 - \exp(-\lambda_w t_g)}{\lambda_w} \cdot V_{dep} \cdot a_i(t) \quad (52)$$

Here $p_i^{dust}(t)$ is the activity concentration of radionuclide i in the edible portion of the plant resulting from foliar interception of dust (pCi g^{-1}). It is derived by assuming that the concentration of radionuclides in dust is constant over the growing period. The derivation is analogous to that for foliar interception of overhead irrigation.

B.6 “CALCULATION BASES MINING” MODEL (CBM)

CALCULATION OF PLANT RADIONUCLIDE CONCENTRATION

The model calculates the specific activity of a radionuclide r in the plant n , $C_{n,r}$ (Bq kg^{-1} FW), distinguishing between plants which are present on the mine tailings pile itself, and those plants which are in the surrounding area. The model considers three means of plant contamination:

- Root transfer from the upper ground layer and activity deposits (dust, ground particle) on aboveground plant sections (for plants growing directly on contaminated soil);
- Dust deposit on aboveground plant sections (for plant growing in the area but not directly on the contaminated soil); and
- Spray irrigation.

Three plant types are considered: leafy vegetables, non-leafy vegetables and pasture plants. BfS (2010) notes that contamination from spray irrigation and dust deposition should not be considered for root vegetables.

Calculation of specific activity from root uptake for plants grown on the mine tailings: From the representative values of the specific activity for the upper ground layer of the mine tailings ($C_{Bo,r}$; 10 cm for pasture ground, and 30 cm for field and garden ground) the specific activity as a result of root uptake, $C_{D,n,r}$ (Bq kg⁻¹ fw), is given by the following equation.

$$C_{D,n,r} = T_{n,r} (C_{Bo,r} - C_{Bo,r}^U) \quad (53)$$

Here $T_{n,r}$ is the transfer factor (Bq kg⁻¹ (FW) of the plant per Bq kg⁻¹ (DW) of the ground), and $C_{Bo,r}^U$ is the natural background concentration of the radionuclide in the soil. For instances where the only radionuclides present come from the mine tailings then $C_{Bo,r}^U = 0$.

Calculation of specific activity in plants arising from spray irrigation:

The specific activity in the plant resulting from spray irrigation, $C_{B,n,r}$ (Bq kg⁻¹ FW), with water of the concentration of activity $C_{water,r}$ (Bq L⁻¹), is calculated as follows:

$$C_{B,n,r} = (C_{water,r} - C_{water,r}^U) \cdot \frac{W \cdot f_w}{Y_n \cdot \lambda_{eff,Pf,r}} \cdot (1 - \exp(-\lambda_{eff,Pf,r} t_{n,W})) \quad (54)$$

Here

- $C_{water,r}^U$ is the natural background concentration of the radionuclide in water (Bq L⁻¹; for instances where the only radionuclides present come from the mine tailings then $C_{water,r}^U = 0$);
- W is the spray irrigation rate (L m⁻² s⁻¹);
- f_w is the proportion of the activity deposited with the spray irrigation on the plant (-);
- Y_n is the yield or vegetation density (kg (fw) m⁻²);
- $\lambda_{Eff,Pf,r}$ is the effective staying constant for the radionuclide r on the plant (s⁻¹); and
- $t_{n,W}$ is the time in which the aboveground part of the plants is contaminated during the growth period by spray irrigation(s).

Calculation of specific activity in plants which are downwind of the mine tailings: The specific activity of plants arising from being downwind of the mine tailings, $C_{A,n,r}$ (Bq kg⁻¹ FW), which results from the deposit of radionuclide contaminated dust is calculated as follows:

$$C_{A,n,r} = (\dot{B}_r - \dot{B}_r^U) \cdot \frac{1 - \exp(-\lambda_{eff,Pf,r} t_{n,e})}{Y_n \cdot \lambda_{eff,Pf,r}} \quad (55)$$

Here:

- \dot{B}_r is the ground contamination rate, which results from the dry deposit of the radionuclide r bound at dust ($\text{Bq m}^{-2} \text{s}^{-1}$);
- \dot{B}_r^U is the natural background contamination rate which results from the dry deposit of the radionuclide r bound at dust ($\text{Bq m}^{-2} \text{s}^{-1}$; set to zero if no background radiation is considered); and
- $t_{n,e}$ is the contamination time for plants during the growth period (s).

Note that in the absence of a site specific ground contamination rate it is possible to estimate \dot{B}_r using a reference value of the deposit rate v_G (m s^{-1}) and the concentration of activity in the near-surface air bound in dust, $C_{\text{Air},r}$ (Bq m^{-3}), as follows:

$$\dot{B}_r = v_G \cdot (C_{\text{Air},r} - C_{\text{Air},r}^U) \quad (56)$$

The specific activity of plants growing on the mine tailings is then given by $C_{n,r} = C_{D,n,r} + C_{B,n,r}$, and the specific activity of plants growing in the area of the mine tailings is given by $C_{n,r} = C_{A,n,r} + C_{B,n,r}$.