BIOPROTA

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

THEME 2: Task 5:

Application of Biotic Analogue Data

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OCTOBER 2005

FOREWORD

Assessing the impacts of releases of radioactivity into the environment relies on a great variety of factors. Important among these is an effectively justified level of understanding of radionuclide behaviour in the environment, the associated migration pathways and the processes that contribute to radionuclide accumulation and dispersion among and within specific environmental media. In addition, evaluating the consequences of any radionuclide releases on human health relies on the use of appropriate physiological and dosimetric models for calculating doses and risks. Assessment methods have been developed over several decades based on knowledge of the ecosystems involved, as well as monitoring of previous radionuclide releases to the environment, laboratory experiments and other research.

It is recognised that in some cases data for these assessments are sparse. Particular difficulties arise in the case of long-lived radionuclides, because of the difficulty of setting up relatively long-term monitoring and experimental programmes, and because the biosphere systems themselves will change over the relevant periods, due to natural processes and the potential for interference by mankind.

It is also the case that much radio-ecological research has tended to focus on relatively few radionuclides, eg. Sr-90 and Cs-137. Although this research has been relevant to operational effluent discharges and accidental releases, other radionuclides tend to dominate long term impacts as may arise from the migration of radionuclides from solid radioactive waste repositories. Examples include C-14, Cl-36, Se-79, Tc-99, Np-237. The viability of geological disposal concepts and the long-term sustainability of radioactive effluent discharges, together with the safe and effective management of contaminated land and surface stores for solid radioactive wastes can only be considered in the light of a good understanding of the environmental behaviour of such longer-lived radionuclides. However, the number of radionuclides involved is relatively small, and the number of important processes associated with migration and accumulation in the biosphere, and the related radiation exposure of humans and other biota, is also relatively limited.

The International Atomic Energy Agency's BIOMASS Theme 1 has provided a basis for identifying, justifying and describing biosphere systems for the purpose of radiological assessment. The development of conceptual and mathematical models has been set out and a protocol developed for the application of data to these models. However the BIOMASS Project did not address the details of uncertainties arising from weaknesses in the information base.

BIOPROTA Concept

BIOPROTA provides a forum to address uncertainties in the assessment of the radiological impact of releases of long-lived radionuclides into the biosphere. The programme of work carried out under the auspices of BIOPROTA focuses on those key radionuclides and the various biosphere migration and accumulation mechanisms relevant to those radionuclides. 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 support a transparent and traceable basis for the choices of parameter values as well as for the wider interpretation of information used in assessments.

The BIOPROTA Project up to December 2004 has been managed and supported financially by:

Organisation	Representative	Role of organisation	Website
Agence Nationale pour la Gestion des Déchets Radioactifs (ANDRA)	Elisabeth Leclerc-Cessac	ANDRA is responsible for the management of radioactive waste in France.	www.andra.fr
Empresa Nacional de Residuos Radiactivos, S.A. (ENRESA)	Julio Astudilio	ENRESA is responsible for the management of radioactive wastes generated in Spain and the decommissioning of nuclear power plants.	www.enresa.es
Nexia Solutions Ltd (formerly BNFL Research & Technology)	Mark Willans	Nexia Solutions is a UK BNFL subsiduary company providing technology solutions and services across the nuclear fuel cycle.	www.nexiasolutions.com
United Kingdom Nirex Limited (Nirex)	Paul Degnan	Nirex is the radioactive waste management agency with responsibility to develop and advise on safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials in the UK.	www.nirex.co.uk
Nuclear Waste Management Organization of Japan (NUMO)	Shigeru Okuyama	NUMO is the implementing body for the final disposal of vitrified high-level waste packaged from the spent fuel reprocessing plant. It is a government approved organization responsible for identification of a disposal site, and for the construction, operation and maintenance of the repository, closure of the facility, and post-closure institutional control.	www.numo.or.jp
Posiva Oy	Ari Ikonen	Posiva is responsible for the management of disposal of spent fuel produced in power reactors in Finland, including siting, licencing, construction and operation of the repository.	www.posiva.fi
Svensk Kärnbränslehantering AB (SKB)	Ulrik Kautsky	SKB is responsible for management of Swedish radioactive waste, planning of waste repositories, waste logistics and site selection, including safety analysis, research and development of methods.	www.skb.se

Since January 2005, the Project has been additionally managed and supported financially by:

Organisation	Representative	Role of organisation	Website
Electricité de France (EDF)	Carine Damois	EDF is the main producer of electricity in France. The Laboratoire National Hydraulique et Environnement (LNHE) department works on migration of pollutants in the ground, waste management, water quality, soil contamination, ecotoxicology, ecology, microbiology, health risk assessment, but also fluvial and maritime hydraulics, resource management, industrial flows and combustion, meteorology and air quality.	www.edf.fr
Korea Atomic Energy Research Institute (KAERI)	Yong Soo Yong-Soo Hwang	Kaeri is developing the Korean reference concept for permanent disposal of high-level radioactive waste including spent nuclear fuel and assessing the long term post-closure safety and repository performance.	www.kaeri.re.kr
National Cooperative for the Disposal of Radioactive waste (Nagra)	Frits van Dorp	Nagra has more than 30 years experience in the development of disposal concepts for all categories of radioactive waste. Over the years, Nagra has built up extensive technical know-how and has applied this in site characterisation and performance assessment of deep geological repositories.	www.nagra.ch
Nuclear Research Institute Rez (NRI)	Ales Laciok	In the Czech Republic, NRI is the research, development and engineering organisation responsible for the development of nuclear power technologies, utilization of radionuclides and radiation in industry and medicine, and with a role to undertake fundamental research to support the long-term management and disposal of radioactive wastes.	www.nri.cz

The BIOPROTA output is made available for use of others, but the participants and supporting organisations take no responsibility for the use of the material.

General Objectives

Overall the intention is to make available the best sources of information to justify modelling assumptions. Particular emphasis is 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 project 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. Such solutions may readily take account of the BIOMASS Theme 1 Data Protocol, among other things.

The modelling assumptions considered include the treatment of various features, events and processes (FEPs) of the systems under investigation, the mathematical representation of those FEPs and the choice of parameter values to adopt within those mathematical representations.

The work programme has been organised in three themes:

Theme 1: Development of a Specialised Data-Base for Key Radionuclides and Process Data
Theme 2: Modelling Testing and Development Tasks
Theme 3: Site Characterisation, Experiments and Monitoring.

A full list of all the reports that have been produced under each theme is available from the BIOPROTA website (www.bioprota.com).

Objectives of the Natural Analogues Task

The objective of Task 5 within Theme 2 has been to investigate and highlight the scope for the application of natural analogues to support modelling assumptions for dose assessment models relevant to long term releases of radionuclides to the environment.

This report has been prepared within the BIOPROTA work programme. The supporting organisations have agreed that BIOPROTA reports will be printed by those organisations in their normal report series. In this case, Nirex is supporting the printing of this Task report, to make it available for a wide audience. Nirex supports the work of BIOPROTA, but does not necessarily endorse the output. Any questions concerning this report should be directed towards the contributors. The report can be obtained directly from Nirex; it is also available in pdf form at www.bioprota.com along with the other BIOPROTA reports.

Recommended Citation

BIOPROTA (2005). Application of Biotic Analogue Data. A report prepared within the international collaborative project BIOPROTA: Key Issues in Biosphere Aspects of Assessment of the Long-term Impact of Contaminant Releases Associated with Radioactive Waste Management. Main Contributors: E Kerrigan (Task Leader), G Smith, M C Thorne. Published on behalf of the BIOPROTA Steering Committee by United Kingdom Nirex Ltd, Oxfordshire.

EXECUTIVE SUMMARY

Analogue information can increase our conceptual understanding of long-term repository and disposal system behaviour, as well as behaviour of radionuclides in the environment, and thus support post-closure performance assessment (PA). The study of analogues can also provide quantitative data for PA models and a communication method to pass on information to a non-specialist audience.

The EC 5th Framework project NANet has been reviewing past and present use and understanding of natural analogues with the intention of promoting considered application of them in future safety assessments and public communication. The focus of NANet was the near field, far field, and geosphere-biosphere interface, not the biotic environment.

Task 5 within Theme 2 of BIOPROTA has been developed to mirror the NANet project and review biotic analogues that have been, or might be, used in PA. The analogues of interest are those that may illustrate the movement and behaviour of radionuclides in the biotic terrestrial environment as could occur from a repository release.

As with all BIOPROTA tasks, the focus is on a small number of key radionuclides that are potentially important for deep repository safety studies. They are: C-14, Cl-36, Se-79, Tc-99, I-129, Np-237, and the U-238 series. Key parameters related to radionuclide uptake and transfer to, and exposure of, flora and fauna are of significance to this study. Various parameters are used to describe such movement, in particular: K_d, root uptake factors, plant concentration factors, animal transfer factors and crop interception.

This brief overview of the use of analogues in assessments of biotic transfers and accumulation pathways shows that there are some processes for which biotic analogues have been used. However, few long-term radiological performance assessments acknowledge the use of biotic analogues and it appears that relatively little of the potentially available information has been applied.

The continuing aim within BIOPROTA is to identify the qualitative and quantitative information derived from biotic analogue studies and to make recommendations for how this information may be used in future performance assessments. Some significant gaps in knowledge and application of analogue studies are identified and recommendations made for how the situation can be improved and whether there is a need for future studies. However, this review is limited in scope and, in the nature of the BIOPROTA forum, is intended primarily to provide a basis for discussion and to act as a pointer for what further work could be useful at the site-specific level.

To focus future efforts, updated advice would be useful, based on recent project specific assessments, on those processes and data that are both important and relatively poorly understood and are, therefore, potential targets for future analogue studies.

1. INTRODUCTION: SCOPE AND OBJECTIVES

Development and application of Performance Assessment (PA) models often requires information on radionuclide behaviour in conditions that are difficult or impossible to study. Experiments may sometimes be used to justify modelling assumptions in the absence of site-specific data, but these may not always be adequate, due to the long time-scales or large environmental systems that are involved. In such cases, study of systems which present conditions analogous to those under investigation in the PA can be used to provide improved system understanding and values for model parameters for which more directly relevant data are not otherwise available. An obvious example is the behaviour of natural radionuclides in the environment, such as U-235, which could also be released from a radioactive waste repository. Many performance assessments of the deep geological disposal of radioactive wastes have been published and have made use of natural and other analogue information [IAEA, 1999].

Analogue information can increase our conceptual understanding of long-term repository and disposal system behaviour, as well as behaviour of radionuclides in the environment, and thus provide support to post-closure performance assessments. The study of analogues can also provide quantitative data for use in PA models and a method for communicating information to a non-specialist audience [Miller *et al.*, 2000]. For consideration of analogues used at a specific site, see Simmons [2003], who discusses the application of analogues at the Yucca Mountain site in the USA.

The European Community (EC) 5th Framework project NANet is reviewing past and present use and understanding of natural analogues with the intention of promoting considered application of them in future safety assessments and public communication – www.enviros.com/zztop/nanet/nanetmain.htm. The focus of NANet is the near field, far field, and geosphere-biosphere interface, not the biotic environment.

Task 5 within Theme 2 of BIOPROTA has been developed to mirror the NANet project and discuss biotic analogues that have been, or might be, used in PA. The analogues of interest are those that may illustrate the movement and behaviour of radionuclides in the biotic terrestrial environment as could occur due to releases from a closed radioactive waste repository.

1.1 Objectives

The aim of this Task has been to discuss and identify the types of qualitative and quantitative information that can be derived from studies that provide analogue information on element and radionuclide behaviour in the biotic environment and to make recommendations for how this information may be used in future PAs.

The analogues of interest are those related to key radionuclides located in a terrestrial environment that is broadly comparable to the environments in which a repository could be sited. There are a relatively small number of radionuclides that might be expected to be released from a deep geological repository and reach the surface environment. Thus, the report focuses on a sub-set of such key radionuclides, specifically C-14, Cl-36, Se-79, Tc-99, I-129, Np-237, and the U-238 series. Key parameters of significance to this study are soil-water distribution coefficients (K_d values), root uptake factors, plant concentration factors, animal transfer factors and crop interception factors. If no relevant radionuclide analogues exist for the relevant radionuclides, there may be an interest in other radionuclides or elements, whose behaviour is thought to be similar in those relevant environments.

The following Sections examine these issues and Annexes present example reviews of particular analogues relevant to selenium, technetium and uranium.

1.2 Scope

Issues to consider include:

- existing analogues involving biotic transfer and accumulation pathways, e.g. soil-plant-animal pathways;
- current and future exposure pathways, taking into account the potential for exploitation of resources in different environmental conditions;
- human interactions with future biosphere systems as they change;
- radiation doses that arise from natural radionuclides, noting their current concentrations in environmental media and fluxes through biotic media;
- relevance of the analogue information to the systems under study in PAs; and
- scope for development of possible new biotic analogue studies.

Examples of analogues that have been considered for non-biosphere aspects of PA in the past, and could be applied to biotic components of the environment are:

- uranium mill tailings heaps, for example, an evolving biosphere growing around a stream from a uranium mine;
- British Geological Survey soil, sediment, and water maps of Cumbria that have been used to try and trace sources of minerals, including uranium;
- freshwater lakes that were once shallow seas; and
- anthropogenic sources:
    - environments surrounding nuclear power plants and reprocessing plants, e.g. I-129 discharge data, for example from Karlsruhe, La Hague and Sellafield;
    - uptake of Chernobyl releases into flora and fauna;
    - releases from existing waste disposal facilities.

This review is limited in scope and, in the nature of the BIOPROTA forum, is intended to provide a basis for discussion and act as a pointer for what further work could be useful at the site-specific level.

2. WHY ARE ANALOGUES USED?

The study of systems which present analogous conditions to those under investigation in a PA can be used to provide improved system understanding and values for model parameters for which more directly relevant data are not otherwise available. This simple statement has two corollaries that seem obvious, but need to be remembered when discussing analogues.

Firstly, analogues provide no benefit if there are no data for the analogue either. For example, stable cerium may be considered a reasonable analogue for curium, but this is not helpful if there are no data for stable cerium in the relevant context.

Secondly, one can never be sure exactly how relevant or suitable any specific analogue is to the situation of interest. An analogue could only be proven to be valid by comparing its behaviour in the conditions of interest with that of the chemical species for which it is an analogue. However, if the data existed for that species in those conditions, then there would be no need to use an analogue. Hence, while confidence in the validity of an analogue will increase as the quality of the justification increases, there will always be some residual uncertainty. One aspect of this justification is a clear understanding of the biosphere model for which data are being sought and the assessment context in which it is being applied [IAEA, 2003a].

As with any other choice of parameter values for modelling, decisions on using analogues must take account of the assessment context and particularly the level of realism or conservatism of the assessment. Analogues can be used in PAs for a number of reasons:

- Conceptual models of repositories and their environs illustrate how a site may be influenced by changing climatic, ecological and hydrogeological conditions over long periods of time. Analogue studies are important for scientists and policy makers charged with developing long-term solutions for radioactive waste management, in that they provide insights into, and validation for, many of the concepts and processes used in their predictive models [McKee and Lush, 2004].

- Mathematical models provide a quantitative representation of our conceptual understanding of system behaviour. The mathematical representation of processes can be supported by analogue information.

- Mathematical models do not always inspire the confidence that direct evidence can. It is sometimes difficult for the public to understand the concepts involved. Analogues can, therefore, be used to provide information to a broad audience, including non-technical members of the public. They can create familiarity with complex scientific processes and long timescales.

- Analogues can justify why a process has been considered, because they can represent system behaviour and the surrounding environment over appropriate timeframes and in suitable settings.

- Analogues can be applied quantitatively or qualitatively depending on their purpose of application. They can provide information about the occurrence of, or constraints on, various processes, or determine the conditions under which a process occurs; the effects of the process; or the magnitude and duration of the process.

- Analogues can be used to fill data gaps where specific information is lacking for model parameter values.

- Analogues can be used to validate model assumptions by providing direct data or as alternative evidence to corroborate assumptions relating to the occurrence and characteristics of particular processes or combinations of processes, e.g. where bioaccumulation has previously been estimated on a less technically justified basis. Typically, that basis will have included cautious assumptions, leading potentially to an over-estimate of impacts.

3. TYPE OF ANALOGUES USED

There are several types of analogue, each of which has advantages and disadvantages relative to the others. When undertaking a PA or developing or applying a biosphere model, the assessor must be careful to use the best analogues in context and include all relevant caveats and justification for the data. There may be multiple analogues of differing quality that could be included in a model and the user must assess which are most appropriate. Many analogues that have previously been used to aid understanding of the issues associated with long-term management of radioactive waste have focussed on the near field and far field, including geological transport and retardation, and natural or man-made barriers.

The advantage of natural analogues over short-term laboratory experiments is that they enable the study of actual ecosystems that have evolved over the relevant ecological timescales. The disadvantage is that the boundary conditions and source term are often unknown or poorly controlled. No one analogue would represent all aspects of a repository system, but different combinations of features, events and processes can be observed in nature. In these circumstances, analogue studies can be useful not only in exploring the effects of individual processes on appropriate length and time scales, but in determining how various processes, each with its own characteristic time and length scales, interact. Therefore, the main types of biotic analogue used in performance assessments are:

- analogue isotopes – use of the value of the same parameter obtained for another isotope of the same element (e.g. using data obtained for a stable isotope in place of a radioactive one);
- analogue elements – use of the value of the same parameter obtained for another element (i.e. using data obtained for one element and applying it to another element);
- analogue media – use of the value of a different parameter obtained for the same element (e.g. using an element specific soil–plant transfer factor obtained for one crop and applying it to a different crop); and
- Overall system behaviour and human interaction analogues e.g. system behaviour in the arctic today may be representative of conditions in the UK in the far future, subject to climate change.

Often analogues of one or more of the above types are used in a biosphere model or PA, but the use of an analogue is not explicitly acknowledged, being hidden in the parameter values adopted.

3.1 Analogue Isotopes

In biosphere assessments[1], analogue isotopes are a commonly used form of analogue, and are often used without any specific justification, or even recognition that an analogue is being used. Typical examples include:

- Short-lived fission products whose environmental behaviour has been extensively studied in the context of reactor accidents or routine discharges

1 This term is used to mean assessment of radiation doses and risks following release of radionuclides into the biosphere. There is no single definition of the biosphere, but it is generally taken to mean either the biota themselves including humans, or the environment(s) in which they live and obtain their sustenance.

(e.g. I-131, Cs-134/137) used as analogues for long-lived isotopes of the same elements of relevance for solid waste disposal (e.g. I-129, Cs-135);

- Similarly, short-lived (and readily available) tracer radioisotopes used in experiments are substituted as analogues for the longer-lived radioisotopes found in radioactive discharges or solid wastes. Short-lived radioisotope and stable element data can be integrated as multiple partial analogues, for example, data are available for selenium forage-cow-milk transfer factors based on stable element data and tracer studies using Se-75 as selenous acid;

- Stable isotopes whose environmental behaviour has been extensively studied in the context of chemical toxicity (e.g. Be, certain heavy metals) used as analogues for less common (and less studied) radioactive isotopes found in radioactive discharges or solid wastes. Thus, stable selenium data have been used directly to estimate selenium transfers to animal tissues at equilibrium [Thorne, 2003];

- Naturally occurring isotopes (stable or radioactive) whose observed behaviour in the environment is used to provide an analogue for the behaviour of the same or other isotopes released to the environment in radioactive discharges or from the disposal of solid radioactive wastes.

There are three special issues that could affect the validity of this type of analogue:

- The timescales for experiments or observations on short-lived isotopes may be limited by radioactive decay and so the observations might not reflect all aspects of environmental behaviour in the long-term.

- The chemical speciation of the isotope being considered as an analogue has to be taken into account. Thus, for example, natural uranium present in minerals in soil may behave very differently from an isotope of uranium added to the same soil via the addition of contaminated well water.

- Similar issues arise when a natural source of a radionuclide is considered as an analogue for the behaviour of the same radionuclide released from a solid waste disposal facility. For example, C-14 released into soil from below as a gas will not have the same environmental behaviour as cosmogenic C-14 produced in the above-ground atmosphere.

Research papers deriving basic data from laboratory or field experiments are generally explicit about the isotope(s) to which the results relate, and in some cases about the validity of extrapolating those results to other isotopes. However, most compilations and reviews of data for modelling purposes appear to take as a starting assumption that environmental transfer parameters are element-dependent rather than nuclide-dependent. Furthermore, such compilations and reviews do not always take into account the context in which the data were acquired relative to their intended application.

The assumption that transfer parameters are element, but not isotope, dependent is probably a reasonable simplification in the context of all the other uncertainties in assessment modelling, but nevertheless for most radionuclides it is good practice to give preference to data derived for the specific isotope of interest over data derived for other isotopes. Note for example, that conclusions on I-131 behaviour and activity levels may not apply to I-129 because the mass of iodine associated within a given activity amount of I-129 is millions of times greater than for I-131, so that observations of I-131 may be independent of chemical saturation effects, but the same activity level of I-129 would not necessarily be independent.

However, this is not always true. Studies of technetium animal transfer factors often use the short-lived gamma-emitting radioisotopes Tc-95m and Tc-99m. There are considerable observed differences between observed transfers of these radioisotopes and of Tc-99 and the validity of tracer experiments has been questioned. However, Tc-99 is usually present in the environment at low mass concentrations, therefore experiments involving large masses of Tc-99 should be viewed with caution and it may be more appropriate to use data for Tc-95m and Tc-99m, which are invariably present at low mass concentrations, even in experimental contexts.

3.2 Analogue Elements

The chemical properties of elements follow fairly well established patterns that can be used as a basis for identifying potential analogues:

- Elements in the same group (column) of the periodic table usually exhibit similar chemical behaviour, because they have the same number of outer electrons available to form chemical bonds. This is particularly true of those elements at the sides of the periodic table which form predominantly ionic bonds; and

- Transition elements in the same period (row) of the periodic table also tend to be chemically similar to one another. This is especially true of the lanthanides and actinides.

However, chemical similarity does not necessarily translate into similar behaviour in the environment. For element analogues within the same group of the periodic table, the differences are often large: for example, strontium isotopes have masses about twice those of calcium isotopes and this significantly affects their environmental behaviour relative to calcium. For period analogues, such as the lanthanides, the mass differences may be much smaller. However, the ionic radius and electronic configuration of such period analogues can influence behaviour more than mass. Thus, for example, over the lanthanide series the predominant environmental oxidation state changes from 2+ to 4+ as the atomic number increases, with concomitant changes in environmental behaviour.

It follows that, although elemental analogue information can be helpful, especially if no other information is available, it is appropriate to understand how these other factors may influence the validity of the analogue.

3.3 Analogue Media

3.3.1 Different Soils

The sorption coefficient or K_d value, is a measure of the partitioning of a contaminant between solution and solid. It can be estimated in the laboratory or the field. However, the K_d value is a function of many physical and chemical characteristics of the solid and the liquid. Soil K_d values can vary significantly with soil type, and the major sources of such data (e.g. Sheppard and Thibault [1990]) provide different reference values and uncertainty ranges for different soil types. For shorter-term assessments (routine discharges, accident consequences) it is generally recognised that K_d values need to match the soil types of interest. For longer-term assessments for solid waste disposal, approaches vary: in some cases (e.g. the ANDRA approach), the present-day soil type is assumed to persist for at least the duration of the current climatic state (i.e. temperate conditions), and then to evolve in response to climate change (e.g. changing to a cryosol as the climate cools towards a glacial episode); others use the uncertainty about future soil conditions as an argument for adopting generic soil K_d values, which may be ‘averages’ over soil type, those for the soil type expected to maximise doses, or simply the type for which data are most readily available.

Another type of analogue that is often applied unknowingly, or at least without detailed justification, is the use of soil–plant transfer factors for soil types different from that for which the factors were derived. Many authors stress the dependence of plant uptake on soil type for some radionuclides, for example nickel – and particularly the need for assumptions about sorption on the soil type to match assumptions about uptake (a high soil K_d is less likely to be associated with high uptake by plants). However, many compilations of soil–plant transfer data make little or no reference to the soil type for which the values were derived. This may be defensible in the context of the relatively high uncertainties associated with a long-term assessment for solid waste disposal, but it is less so for discharges or accidental releases.

3.3.2 Different Plant Types or Animal Products

The other common use of analogues is to assume that the concentration factor for a given element in a plant type or animal product for which data are not available will be the same as for the same element in a plant type or animal product that has been studied. Clearly, the validity of such analogues depends not only on the similarities in nature between the different plant types or animal products but also on the chemical behaviour of the element in question. In general, this type of analogue will be more appropriate for elements that tend to distribute evenly in the environment than for those that demonstrate strong affinity for certain parts of plants or animals. Even with widely distributed elements, careful consideration would be needed of each specific case.

In the absence of specific data on an animal type of interest, often another animal that is similar is used; for example, in the absence of specific data for ducks, data for hens are used, and the same is true for sheep and cows. Often, data for laboratory animals such as rats or Japanese quail are used, and the data are then inferred to be applicable for similar animals. There are problems with this approach, but the application of allometric techniques to predicting radionuclide tissue concentrations [Higley *et al.*, 2003] may confirm such analogues provide acceptable derived data.

Some analogues are relatively obvious (although being obvious does not always mean they are correct), e.g. root vegetables and potatoes, grass and leafy green vegetables (for irrigation interception etc). Other analogues might seem obvious, e.g. between products from the same type of animal, but need to be approached with caution, especially when one product (e.g. milk, eggs) is collected during the life of the animal whereas another (e.g. meat) arises only when the animal is slaughtered. Uptake of radionuclides into animal products is determined by function, for example, iodine uptake by the thyroid between species is more similar than iodine uptake by the thyroid and meat in a particular species.

3.4 System and Human Interaction Analogues

The present is often thought to be the best analogue for the future. This is particularly true when making judgements on human behaviour to include in a PA.

Many assumptions are made regarding the work, rest, inhalation and consumption habits of critical groups, as is apparent in OCRWM [2003] where the dietary, lifestyle and dosimetric characteristics of the human receptor to be assumed for the

Yucca Mountain repository are documented. This is consistent with the licensing rule at 10 CFR Part 63, whereby a hypothetical person called the Reasonably Maximally Exposed Individual (RMEI) represents the potentially exposed population.

The RMEI, as with many critical groups, applies the same habit assumptions in the future as in the present. Some human behaviour will change over time due to external influences, for example, climate change affecting farming and animal husbandry practices, time spent outdoors, *etc*. To represent such changes, present-day data from locations with climates or other features analogous to those expected at the site of interest in the future could be used.

There are also assumptions made regarding Reference Man [ICRP, 1975 and 2003; IAEA, 1998]. Until the publication of IAEA [1998], the physiological data assumptions for an Asian man were the same as for a Caucasian population of Europe and North America. Such an assumption has implications for the radiation dose received.

The NRPB [Smith and Jones, 2003] have summarised default habit data currently used at the NRPB for generalised radiological assessment purposes and ANDRA has defined current critical group behaviour as an analogue for future potential exposure groups.

Climatic and regional analogues for analysis of biosphere systems under environmental change have been used. Dynamic evaluation of the effects of climate change has been considered within the BIOCLIM project [BIOCLIM, 2004].

4. PARAMETERS FOR WHICH ANALOGUE INFORMATION COULD BE OBTAINED

4.1 General Considerations

There have been a number of studies measuring global fallout in the environment and related tracer radionuclides have been used to enhance knowledge of biogeochemical processes and food chain pathways. These processes control the transport and accumulation of trace substances in the environment. Radionuclides can be easily measured in environmental media and biological tissues at mass concentrations that are not harmful to the organism and do not perturb normal biogeochemical processes [Whicker and Pinder, 2002]. This information can then be used to determine behaviour patterns in similar ecosystems.

However, it should be noted that the particular release mechanisms into the environment may affect the relevance of an analogue. For example, transfers of technetium in soil after atmospheric dispersion of the pertechnetate may have little relevance to the transfers of technetium in soil that would occur as the result of its entry in groundwater from below through a reducing zone.

C-14, Cl-36, Se-79 and I-129 are all isotopes of elements of substantial environmental importance and the behaviour of the stable element in each case is relevant to the behaviour of the radionuclide. For Tc-99, the situation is very different because there is no stable element to use as an analogue. As pertechnetate, there are useful analogies to iodide, but there are limitations. For example, pertechnetate is taken up by the thyroid in a manner very similar to iodide, but because it cannot be used to make thyroid hormones, it is rapidly lost (unlike iodine) and is not present in tissues in organic form. Also, the redox sensitivity of technetium chemistry impacts its behaviour. Redox sensitivity is also an issue for Se-79 and I-129.

For Np-237, it is probably best to rely on the relatively extensive radionuclide-specific environmental data that are available and to avoid analogies, as the chemistry of neptunium is quite distinct from that of the other actinides. However, it can be useful to compare environmental behaviour between Np, Pu, Am and Cm, but only after each has been analysed separately.

For the U-238 series, some reliance can be placed on data for naturally occurring uranium and its radioactive progeny; see for example the substantial literature in Annex A of UNSCEAR [2000]. However, there are caveats arising from distinctions in the chemical form of the element present in the environment or postulated to be released from a repository.

4.2 Migration and Behaviour of Radionuclides in Soil

Migration of radionuclides in soils is more complicated than the simple leaching process typically considered in assessment models. For example, assumptions about mixing in the surface soil may need to allow for biotic disturbance including ploughing by man. Empirical data for the rate of infiltration of weapons test fallout implicitly take account of all the processes on-going at a site, so use of such data provides an integral representation of all the processes of relevance, provided that the site of interest and the analogue site are suitably matched. A recent review of soil processes and how they are included in a variety of models for radionuclide accumulation in soil is provided in BIOPROTA (2005a).

4.3 Soil – Water Distribution Coefficients - K_d

K_d values, although usually assumed for modelling purposes to be constant for a given system under specified conditions, vary over many orders of magnitude for different situations. Fortunately, in most situations involving radionuclides, the mass concentrations are low and K_d values are normally independent of the radionuclide concentration [Leung and Shang, 2003]. However, soil type is very important in the determination of K_d; for example, Sheppard and Thibault [1990] found K_d values of between 1 and 70 l/kg for iodine under four different soil type conditions. However, the inferred dependence on defined soil types may be a reflection of more fundamental considerations that are amenable to study. It is suggested that soil K_d values are better predicted by a small number of cofactors which depend on the element in question. These may include aspects of soil type (e.g. organic content) for some elements, but may also include factors such as the pH and/or Eh of the soil and the concentration of competing elements in the soil (e.g. potassium content is a strong cofactor for caesium K_d in soil).

Because soil K_d data exhibit large variations between different soil textures and even on small spatial scales, analogue K_d data from a specific site may not be more appropriate than generic data. This implies that site-specific data should not be used to the exclusion of other available data. Even dramatically different soil types can be expected to exhibit overlapping distributions of K_d values [Sheppard, 2005].

When considering a choice of analogue for K_d, it is useful to consider at the same time the factors relevant to assumptions about root uptake, since sorption and root uptake may be anti-correlated.

4.4 Root Uptake

There are many studies of uranium mill and mine tailing sites that discuss the behaviour of uranium-series radionuclides in the environment [Krizman *et al.*, 1994]. The Brodueirs and Madruga [2001] study quantified the availability of Ra-226 in soil for plant uptake at a site where a large amount of solid waste from uranium ore treatment had accumulated in dams. To reduce the dispersion of radionuclides in the environment, some dams were vegetated with pine trees and shrubs. The subsequent measurement of Ra-226 in the waste and transfer to plants gave an indication of its bioavailability. The study found that the concentration ratio depended on the Ra-226 concentration in the waste. This information can be used to determine the concentration ratio in the same or similar types of plants in soils that have a similar source term of uranium.

The lanthanides are generally considered to be analogues for actinides, provided they have the same oxidation state. Knowledge of lanthanide behaviour in the environment may provide an insight into the behaviour of the transuranic elements. As an example, a study of freshwater plants, molluscs, surface water, porewater and sediment [Heidenreich and Weltje, 2002] could provide analogues for the behaviour of radionuclides released from a repository upon entering a freshwater ecosystem. This study was conducted in the Netherlands and may be an appropriate analogue for freshwater ecosystems in sites where the sediment profile is similar.

A study of the site of a disused uranium mine located in the Extremadura region of southwest Spain looked at the transfer factors for natural uranium (U-238 and U-234), thorium (Th-232, Th-230 and Th-228), and Ra-226 isotopes between plants (pasture) and granitic and alluvial soil samples [Vera Tome *et al.*, 2003] (details are provided in Annex 3). A high degree of variability in transfer factor values was obtained for thorium isotopes, because Th-228 is difficult to measure due to the effects of Ra-228 decay within the plant samples. Factors such as soil characteristics, climatic conditions, plant type, part of plant concerned, physical-chemical form of the radionuclides, and the effect of competitive species can influence the transfer factor values. However, the data obtained for transfer to plants from Mediterranean soils can be used to confirm (with caution) transfer factor values used in models where the soils are of a similar composition to those in the study.

Flowering cabbage, the vegetable with one of the greatest values of soil to plant transfer factors, could be used as a bio-monitor for the radioisotope contamination in vegetables [Leung and Shang, 2003]. Such plants, used in phyto-remediation, may provide an upper estimate of potential root uptake factors, but are not relevant to predicting typical uptakes into crops consumed by man or animals, but they may be relevant in some circumstances

4.5 Translocation Factors

Translocation is defined [ICRU, 2001] as the transfer of materials from one part of a plant to another. This quantity is used to estimate the activity density in an unmeasured tissue from another tissue in the same plant which has been measured. More precisely, the translocation factor is the mass activity density (Bq/kg) in one tissue, typically an edible tissue, divided by the mass activity density (Bq/kg) in another tissue of the same plant or crop. This definition is relevant to, for example, estimating the radionuclide content in root crops or seeds, based on information on the radionuclide content of the leafy part of the plant.

The translocation factor also can be defined [ICRU, 2001] as the mass activity density in the edible tissue divided by the activity contained on the mass of foliage covering a square metre of land surface. In this case, the purpose is to support assessment of the transfer from the plant surface (eg following wet or dry deposition) into the plant, where it cannot be subject to weathering or removal by washing. Further translocation, as defined above, can then occur.

In both cases, the transfer factor for a specific radionuclide varies with chemical form, the timing of measurements, species, growth stage and nutrition status of the plant [Voigt *et al.*, 1991]. Translocation is not well distinguished for different plant types. Crops are sometimes divided into those for which the exposed above ground parts are edible (green vegetables, pasture, most fruits) and those for which the edible parts (either by man or animals) are clearly separate and/or below ground (grain, root vegetables and potatoes), but the available data are rarely good enough to distinguish even between these two groups.

Given lack of good data and variations in the way that relevant translocation processes are modelled, special care needs to be taken in understanding the assessment model when adopting analogue assumptions for this parameter. It is noted for example, that within a recent model comparison and testing exercise, the biggest reasons for discrepancies in results for assessments of concentrations in foodstuffs were associated with the treatment of weathering and translocation, post deposition. Values for translocation varied considerably between the participants and there were different interpretations of weathering and translocation data [BIOPROTA, 2005b].

4.6 Animal Product Transfer Factors

Thorne [2003] has reviewed animal transfer factors (TFs) for radioactive isotopes of iodine, technetium, selenium and uranium.

Transfer factor values for radionuclides to milk from cows, sheep and goats are generally assumed to be comparable, given a general lack of species-specific data. However, IAEA [2003] illustrates at Annex CIII that where specific data are available, in this case distinguishing I-129 uptake into sheep milk as opposed to cow's milk, this may have a notable influence on the estimated dose.

There are few data available for chickens or other poultry. Transfer factors for duck eggs, however, have sometimes been assumed to be similar to values for chicken eggs [Thorne, 2003].

Depending on the element, when transfer factors for poultry are completely lacking, an allometric method can be used to scale values for cattle tissues by multiplying this value with the inverse ratio of the body masses, which is approximately equal to 3/500. For example, according to Thorne [2003], there seems to be a negative correlation between body mass and iodine transfer factors for various species, even though chickens do seem to be an exception.

Data on transfer factors of neptunium are scarce and more research is needed to be able to discuss the processes involved. Also, transfer factors for thorium have received little attention due to the low bioavailability of thorium for uptake into edible animal products. ICRP [1995a], however, concluded that the metabolic behaviour of thorium is similar in many aspects to plutonium. Uranium on the other hand behaves more like calcium once it enters the body, due to similar chemical properties, and hence moderately high transfer factors to eggs are expected, though account needs also to be taken of the limited gastrointestinal absorption of uranium relative to calcium. As discussed in Thorne [2003], it is possible to pull together a wide variety of information on the elements of interest into coherent estimates of ranges of transfer factors for various animals in different animal products.

4.7 Environmental Change

Hibbs *et al*. [2000] examined the occurrence of high selenium concentrations in shallow groundwaters in former marsh areas in Orange County, California. The research showed groundwater discharge to creeks, which contained high concentrations of Se due to immobile forms of Se having accumulated in the sediment at the bottom of the marsh due to anoxic conditions. After transformation to agricultural land, the Se was released by transformation into more volatile forms due to oxygenated groundwater replacing oxygen-deficient waters. This example illustrates the potential for environmental change to result in the acute release of particular chemical species. Although it may not be possible to accurately predict environmental change at a site in the very long-term future, changes at other sites today may provide analogues for future conditions at the site of interest. Changes of interest include for example, those processes which modify the assumed constant equilibrium distribution coefficients adopted in assessing accumulation in soils and sediments.

5. DISCUSSION

This brief overview of the use of analogues in assessments of biotic transfers and accumulation pathways shows that there are some processes in which biotic analogues have been used in long-term radiological PAs, but the scope for further use is apparent.

The continuing aim within BIOPROTA is to identify the qualitative and quantitative information derived from biotic analogue studies, and to make recommendations for how this information may be used in future performance assessments.

The IAEA Programme on Environmental Modelling for Radiation Safety (EMRAS) and more particularly the working group for revision of the Technical Reports Series No. 364 “Handbook of parameters values for the prediction of radionuclide transfer in temperate environments” [IAEA, 1994], is giving consideration to the use of analogues to provide data for the broad range of biosphere transfer parameters where there are currently no data. The aim of the report, due to be issued in 2005, is to propose and justify the use of analogues in missing data cases. This generic work complements the BIOPROTA activity which focuses on the relatively few radionuclides and processes relevant to long-term radiological assessments associated with waste disposal.

Analogues can play many roles within a radiological assessment. They have many advantages as a data source as detailed in Section 2. However, good analogues are rare and they often require careful interpretation.

To focus future efforts, updated advice would be useful, based on recent project specific assessments, on those processes and data which are both important and relatively poorly understood, as these are potential targets for future analogue studies.

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ANNEX A: BIOACCUMULATION OF SELENIUM

Description: Marshland was reclaimed for agriculture in early 1900's. Drainage was created, displacing cattle and sheep grazing from the marsh edges. Deformities in waterfowl in the 1980's were attributed to the toxic effect of selenium bioaccumulation. Field experiments observed an increase in selenium between upstream and downstream sampling locations in the creek. The difference has been attributed to high concentrations in groundwater. The high groundwater concentrations are in areas where marshland existed. Before agriculture, Se was weathered from marine sediments and rocks and transported via run-off to the marsh which was oxygen deficient. The anoxic conditions resulted in immobile forms of Se which accumulated in bottom sediment and were stable until drainage allowed oxic groundwater to disturb the sediments and redissolve Se. Selenium and sulphate follow similar patterns of mobility in oxic and immobility in anoxic environments.

A study area location map, showing the location of the historic marsh region in the Irvine Sub-basin, and upstream and downstream surface water sampling stations on San Diego Creek, Como Channel and Santa Fe Channel is given in Hibbs *et al*. [2000]. A conceptual diagram indicating contemporary inflows of selenium-laden and oxic groundwaters into creeks and channels that have been constructed to maintain the water table beneath the land surface in the Irvine sub-basin and a map showing the selenium concentrations in groundwater at the sample sites are included in this paper as well.

Relevance: The study is relevant for the understanding of the mobility of Se in the zone between the deeper geosphere and the surface environment associated with different environmental conditions and the transport of Se in groundwater and surface water flows. It is an example of a marsh Geosphere-Biosphere Interface involving Se dispersion in sediments.

Limitations: The study is limited to the surface / near-surface migration processes for Se in a temperate environment.

Quantitative Information:

Two tables are included in Hibbs *et al*. [2000] showing the concentration of selenium at upstream and downstream sampling stations, and stream discharge at upstream and downstream gauge stations, respectively.

Uncertainties: Hibbs *et al*. [2000] includes statistical data on the measured parameters.

Time-scale: This analogue is at a historic and human time-scale.

PA/safety case applications: There is no evidence of the analogue having been used in PA/safety case applications.

Communication applications: There are no known communication applications of this analogue.

Reference:

Hibbs B, Lee M, Walker J, [2000]. Selenium remobilization due to destruction of wetlands in Orange County, California. Environmental Geosciences, **7(4)**: 211.

Added Value Comments: The migration data in changing environmental conditions could be useful for validating model migration and accumulation codes.

Potential follow-up work: Information on other factors affecting the selenium concentration in groundwater in this analogue is needed.

Keywords: Marsh; Groundwater; Selenium; Bioaccumulation.

ANNEX B: TECHNETIUM SORPTION ON SEDIMENTS

Description: Uptake of Tc-99 by organic matter may involve prior reduction to Tc(IV) and therefore highly reducing conditions enhance organic uptake of Tc. The low organic content of sediments at Needle's Eye is potentially why Tc is associated with the carbonate phase. Formation of relatively soluble Tc(IV) carbonate may explain low uptake of Tc in iron and sulphate reducing sediments there.

The sites are: Rostilde Fjord, Manager Fjord, Needle's Eye Solway Firth, Esk Estuary.

Coastal environments with reducing waters and/or sediments represent potential sinks for Tc-99 discharged to sea [Keith-Roach *et al.*, 2003]. Technetium-99, which has a long radioactive half life and is sometimes an important radionuclide in environmental safety assessments around nuclear facilities, is present in many discharges from nuclear facilities in its soluble form TcO_4^-. In oxidising conditions, this chemical form is very stable and not very particle reactive. Chemical reactions and micro organisms can change the chemical state of technetium to the reduced, +4, state in which it more particle reactive and can, therefore, be deposited in sediments. Organic matter has also been found to be correlated with Tc uptake in soil.

Keith-Roach *et al.* [2003] investigated whether "*particular biogeochemical processes are predominately responsible for reducing and binding technetium in sediments*". They found that *"organic matter appears to be the most important binding phase for Tc(IV)".* The uptake of Tc by organic matter is enhanced by reducing conditions in the soil, which facilitates the reduction of technetium to its +4 state. Sulphides are important in reducing TcO_4^-, but Keith-Roach *et al.* [2003] were not able to determine whether sulphides are a common binding phase for Tc. In two of the four sites investigated, iron oxyhydroxides appear to have had a significant, but non-dominating, effect in binding Tc to sediments. Two of the sites considered were estuaries near Sellafield in the United Kingdom and two of the sites were fjords in Denmark. These latter were found to be better sinks for Tc than the former.

Keith-Roach *et al.* [2003] includes site descriptions, including redox characteristics, sediment type and major Tc input to sediments. There is also a map with the geographical locations of the sample sites, but this is not very detailed. Water conditions at the fjord sample sites are included.

Relevance: Due to its long half-life and the fact that it is considered one of the most important radionuclides in environmental safety assessments for some nuclear facilities, a thorough understanding of the behaviour of Tc-99 in the environment is very important.

Limitations: There is currently no information available from these sites on mixing within sediment columns. A lack of data on mixing prevents the determination of the source and age of various elements in the sediments. There is also a lot more to learn about Tc-99 behaviour in reducing conditions. Sulphides are not a common binding phase for Tc-99 in the environment. However, where they occur, sulphide-rich environments can be effective in the reduction of TcO_4^-, but more research is required on this topic.

Quantitative Information: Quantitative information on the total Tc-99 concentration of each sediment type considered, and relative uptake factors for biota at the field sites, are given in Kershaw *et al.* [1999], as are results of the sequential extraction of Fe and Tc-99 from sediments at all the four sites considered.

Time-scale: This analogue relates to a human time scale.

PA/safety case applications: UKAEA have considered some Needle's Eye findings for developing natural safety indicators for the "Run 1" safety assessment for a hypothetical LLW repository at Dounreay. Whether specific use has been made of Tc-99 sorption data from any of the sites is not known.

Communication applications: There are no known communication applications of this analogue.

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Added Value Comments: Keith-Roach *et al*. [2003] found that pore water concentrations within the soil column reflected current Tc emissions rather than historical emissions. There is, however, no research to support this hypothesis. However, this conclusion may reasonably be drawn for the oxic upper part of a sediment column.

Potential follow-up work: Further studies on the effects of sulphides and micro organisms on the sorption of technetium on sediments are needed to improve understanding on the behaviour of this radionuclide in aquatic systems.

***Keywords**:* Sorption; technetium; aquatic systems; Sellafield.

ANNEX C: TECHNETIUM BIOAVAILABILITY

Description: A detailed understanding of the behaviour of radionuclides in soil is important to predict, among other features, the bioavailability of nuclides for root uptake by plants. There are a large number of factors that influence the bioavailability of radionuclides in soil, which can play a part for all radionuclides, or for some specific radionuclides.

Tagami and Uchida [1996] looked at the role of micro-organisms in influencing the bioavailability of technetium in soil. They specifically look at their influence under waterlogged conditions, since this represents the condition of paddy fields for rice production. In this system, "*oxidation-reduction reactions play an important role in controlling the mobilization and biological accumulation of some toxic trace elements from soil.*" Tc-sorption on soil or minerals has been reported under relatively reducing conditions generated by microbial and/or chemical reactions. The low redox conditions in rice paddy soil are established by the waterlogging during the cultivation period. Soil samples with different microbial activity levels, managed by adding glucose to the soil sample, were examined and the influence of water content, pH, the redox potential (Eh), cation exchange capacity (CEC), anion exchange capacity (AEC), active-Fe, organic-C, total-C, total-N and active-Al were determined. This is important, since Tc might be sorbed onto organic substances and Fe and Al oxides while passing through the soil. Tagami and Uchida [1996] reached the conclusion that "*low Eh is caused by oxygen consumption of micro organisms and that waterlogging prevents entry of oxygen from the atmosphere.*" Tc is transformed from soluble TcO_4^- to more reduced forms such as TcO_2, $TcO(OH)_2$ or TcS_2 in the presence of micro-organisms under waterlogged conditions, which diminishes bioavailability of technetium for root uptake by plants.

Relevance: Two different soil types were studied in both aerobic and anaerobic conditions, the latter simulating rice paddy field conditions.

Chemical speciation influences bioavailability and plant uptake, and can help the investigation and prediction of other aspects of environmental distributions.

Limitations: The use of information on bioavailability of radionuclides depends very much on the specific conditions under consideration. As for the Tagami and Uchida [1996] article, the information depends very much on soil characteristics, waterlogging and perhaps on the type of micro-organisms present in the soil. Other factors, including temperature and water flow in soil, also limit the applicability of this analogue to different settings.

Quantitative Information:

The Tagami and Uchida [1996] article includes a table with the characteristics of the soil samples considered and a table with correlation coefficients between soil properties and relative concentration (RC) of Tc in the bottom solution after one hour. A plot of relative concentration of Tc-95m in bottom solution after one hour versus carbon content, and the time dependence for relative concentrations of Tc-95m in surface solutions are included in the Tagami and Uchida [1996] article as well. This article also contains six plots showing the time dependence of relative counts of Tc-95m in surface and in bottom-solutions and Eh(V) for different glucose concentrations for two soil samples. The final two figures in the Tagami and Uchida [1996] article show the Tc-95m activity concentration from the surface of two soil samples.

Uncertainties: Uncertainties in determining the contributions of the large variety of factors affecting bioavailability of radionuclides for root uptake by plants are mainly determined by the individual, or combined, effects of the various factors, which can be both hard to measure and difficult to separate.

Time-scale: This analogue deals with a human time scale (i.e. between zero and a hundred years)

PA/safety case applications: There is no evidence that this analogue has been used in PA/safety case applications.

Communication applications: The analogue could be used to illustrate different bioavailability in aerobic and anaerobic conditions.

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Tagami K, and Uchida S, (1996). Microbial Role in Immobilization of Technetium in Soil under Waterlogged Conditions. Chemosphere, **33(2)**: 217-225.

Added value comments:

Potential follow-up work: Research on bioavailability of a larger set of radionuclides in general for a wide range of soil types and in a wide range of settings is on-going and some of this research should be able to build on the type of information given in the Tagami and Uchida [1996] article.

Keywords: Bioavailability; Sorption; Technetium; Micro-organisms.

ANNEX D ANTHROPOGENIC URANIUM SOURCES

Description: Ketterer *et al*. [2003] looked at the procedure of measuring the U-236/U-238 ratio using rapid inductively coupled plasma mass spectrometric (ICPMS) methods. Three sites were considered, Rocky Flats, the Mersey Estuary and the Ashtabula River.

Offsite soils from the vicinity of the Rocky Flats Environmental Technology Site (RFETS), 20 km northwest of Denver were collected. The site was used between 1952 and 1989 as a plutonium fabrication site. Other studies of the site have found elevated levels of U-236/U-238.

Sediments from the Mersey estuary, UK and Ashtabula River, Ohio were used to measure the U-236/U-238 ratio. The estuary contains inter-tidal salt marshes that accumulate radionuclides in sediment profiles. Sellafield-linked increases in the U-236/U-238 ratio have been found by Fox *et al*. [1999].

From 1962 to 1988, the RMI Ti facility fabricated various U metal rods and tube products and releases of U to the watershed of the Ashtabula River have been reported to the US Department of Energy (DOE). A second source of uranium in the river is a titanium ore processing facility which has been in continuous production since the late 1950's. An archived sediment core from the Ashtabula River was selected for determination of the U-236/U-238 ratio. The Ketterer *et a*l. [2003] study identified elevated uranium concentrations and distinguished the contribution of the two sources.

Relevance: The ratio of U-236/U-238 in the environment from man-made sources can be used to determine the potential sources of U-236/U-238.

Limitations: The exact source of the anthropogenic uranium in the analogue is not known and may be as a result of accidental releases, e.g. due to fires and uranium-contaminated waste oil leakage from drums and various waste disposal practices. The U-236/U-238 ratio allows identification of reactor irradiated fuel and the different sources of contamination.

Quantitative Information: ICP mass spectral scans in the mass range (233.8-236.2) for the RFETS site with U-236/U-238 = 0.8 ppm is included. A figure of U-236/U-238 atom ratios, U-238/U-235 atom ratios, and U concentration (ppm) for Ashtabula River sediment from the 1997 sediment core is provided. There is also a scatter plot of U-236/U-238 vs. U-235/U-238 which indicates mixing between crustal and/or Ti ore uranium and non-natural RMI U of multiple isotopic compositions.

A table with U-236/U-238 ratios for samples from the RFETS site is included, which clearly indicates the spatial variability in the U-236 distribution around the site.

Uncertainties: Based upon the analysis of replicates and considerations of possible systematic errors, uncertainties of ±5% are found for U-236/U-238 atom ratios of 1-100ppm. Uncertainties for samples with U-236/U-238 below 1ppm are on the order of ±10%.

Time-scale: The analogue deals with a human time-scale, i.e. between 0 and 100 years.

PA/safety case applications: There is no evidence of the analogue having been used in PA/safety case applications.

Communication applications: There are no apparent communication applications of this analogue.

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Added Value Comments:

Potential follow-up work: Further studies of soil U-236/U-238 ratios, and U-236 inventories, would be needed to better demonstrate the migration of released uranium near RFETS.

Keywords: U-236; Rocky Flats Mersey Estuary; Ashtabula River; uranium; anthropogenic; sediments; Sellafield.