International Union of Radioecology

RECOMMENDATIONS FOR IMPROVING PREDICTIONS OF THE LONG-TERM ENVIRONMENTAL BEHAVIOUR OF ^{14}C, ^{36}CL, ^{99}TC, ^{237}NP AND ^{238}U

Findings of the IUR Waste and Radioecology Task Force

IUR Report 6 : 2006

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[Appendix 4 lists in full the organisations abbreviations]

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[Appendix 4 lists in full the organisations abbreviations]

Executive Summary / Abstract

The International Union of Radioecology (IUR) created a Task Force "Radioecology and Waste" with the overall objective of promoting cooperation between radioecologists for research in the field of radioactive waste management.

This report provides an overview of the available knowledge related to the behaviour of ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np, ^{238}U in both terrestrial and aquatic ecosystems based on information that has been collated by experts through the activities of this Task Force. The findings have been made available to the BIOPROTA group, and collaboration with BIOPROTA has been maintained through the work of the Task Force.

An overview of the behaviour of the studied radionuclides is presented with the help of interaction matrices that were developed for terrestrial and aquatic environments. These matrices identify the main components and processes that describe the behaviour of the radionuclides in these two broad environment types. Furthermore, the potentially relevant processes were identified for each specific studied radionuclide (^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np, ^{238}U).

There are wide differences in data availability and process descriptions between the five studied radionuclides. For ^{14}C there is a good understanding of the behaviour, supported by empirical data, as opposed to ^{237}Np where there are few empirical data and assessments are usually done using data and knowledge from analogue elements, *e.g.* uranium and plutonium. New experimental studies should therefore be initiated to fill these gaps.

General approaches for improvement of the assessments models were also summarised, together with the needs for research and development for each specific radionuclide. Conclusions, recommendations and proposed future work by the Task Force are provided. For example, interaction matrices for other radionuclides (e.g. ^{59}Ni, ^{79}Se, ^{94}Nb, ^{129}I, ^{226}Ra, ^{239}Pu, and ^{241}Am) should be developed, supported by descriptions of the relevant processes and the identification of preferred ways of modelling.

Contents

1. Introduction

The "Radioecology and Waste" Task Force was launched during the Workshop "Mobility in Biosphere of Iodine, Technetium, Selenium and Uranium" organised in April 2002 by the French National Radioactive Management Agency (ANDRA) and the International Union of Radioecology (IUR). The idea was well received by the IUR members participating in the workshop, and seventeen of them decided to join the Task Force. Part of the fees, collected during the workshop, was allocated to the new Task Force.

There was consensus among the participants in the workshop that radioactive waste management issues, and, in particular, the management of High Level Radioactive Waste (HLW) is one of the priority areas in radioecology. It was decided that the overall objective of the Task Force should be to promote cooperation between radioecologists for research in the field of radioactive waste management.

1.1. The IUR Radioecology and Waste Task Force

The Task Force adopted as a working strategy the organisation of workshops focused on key issues, e.g. the behaviour of key radionuclides. The Task Force had 24 members, listed in Appendix 3.

The more specific objectives of the Task Force are to:

- explore processes at the geosphere – biosphere interface and how to treat releases to the biosphere from underground sources;
- improve models of long-term behaviour and accumulation of radionuclides in the environment;
- identify knowledge and data gaps for key radionuclides in the HLW disposal context and propose approaches for filling the gaps. It was decided to start the work of the Task Force with a focus on C, Cl, Tc, Np and U. These elements were identified as important in HLW management by the waste management agencies participating in BIOPROTA (BIOPROTA is an ongoing international project in the field of radioactive waste management involving waste management agencies) and show contrasting behaviour in the environment. Other radionuclides of interest may also be included in the study as the work progresses.
- compare strategies for assessment of the radiological impact of radionuclides released into the environment: deterministic versus probabilistic approaches.

There was consensus that the Task Force should have a scientific profile, i.e. provide a forum for open and informal scientific debate and propose, but not undertake, research or development projects.

One, meeting took place in Monaco in 2002 and three scientific workshops were held from 2002 to 2006, in Merlewood, Madrid and Mol. The list of participants who took part in the meeting and each of the workshops can be found in Appendix 3.

1.2. The Meeting in Monaco

A first meeting of the "Waste and Radioecology" Task Force was occurred during the Conference: "International Conference on Radioactivity in the Environment", held in Monaco in September 2002. Previous to the meeting, a questionnaire was circulated among the members of the Task Force to identify relevant on-going and past research programmes in the field.

It was suggested and approved to organise a first scientific workshop dedicated to the behaviour of ^{14}C, ^{237}Np and ^{36}Cl in the environment. It was suggested to continue close cooperation with the project BIOPROTA, which had been as an initiative of several waste management agencies and the IUR. The following activities were agreed for the first year.

- Creation of Newsletter of the Group.
- Set up an inventory of ongoing research activities in the field.
- Organise a first workshop to take place in 2003 in Merlewood, UK.
- Prepare minutes from the workshop.

1.3. The Workshop in Merlewood

The first scientific workshop of the Task Force, "The Merlewood Workshop", was held at the CEH in the UK and was focused on three key radionuclides in radioactive waste management: ^{14}C, ^{237}Np and ^{36}Cl. During the workshop, interaction matrices were developed to represent the components and processes involved in the turnover of radionuclides in terrestrial and aquatic ecosystems.

One important issue of the workshop was to discuss how the Task Force would work in the future and the cooperation with BIOPROTA. Information on the BIOPROTA project can be found at the project web site: www.bioprota.com.

The members of the Task Force present at the meeting agreed on the following action plan:

- To further develop the interaction matrices discussed during the workshop in consultation with SKB, Nirex and other relevant organisations.
- To prepare a list of existing relevant radioecological and ecological models for the three radionuclides and compile existing literature through enquires to the broader IUR membership.
- To analyse the database of the US registry of workers exposure to transuranic elements with the aim of obtaining information relevant for ^{237}Np and other actinides.
- To prepare a text outlining the view of the group on the use of concentration factors for ^{14}C.
- To suggest possible experimental studies for improving knowledge of ^{36}Cl behaviour in terrestrial environments.
- To carry out a second workshop during 2003, to progress the work carried out since this workshop.

1.4. The Workshop in Madrid

In the first part of the workshop the participants discussed the interaction matrices (for ^{14}C, ^{237}Np and ^{36}Cl) developed during the previous workshop and ranked the identified processes by their significance. In the second part, the group examined two more elements, technetium and uranium. For each of them, the group scrutinised once more the matrices to spot missing relevant processes and to rank the processes represented in the matrices. The third part was dedicated to planning the activities of the Task Force in 2004.

The members of the Task Force present at the meeting agreed on the following action plan.

- To continue the work dedicated to ranking radionuclide transfer processes and identifying knowledge/data gaps with the help of interaction matrices.
- To prepare a third workshop to progress the work carried out.

- To prepare a technical report with the findings of the group on the knowledge and data gaps for the studied radionuclides, suggestions for improvement of the assessment models and research priorities.
- To prepare a technical report with the finds of the group to be published as an IUR Report.

1.5. The Workshop in Mol

The last workshop, at the SCK-CEN (Belgian Nuclear Research Centre), in Mol on the 18th and 19th October 2006, was used to finalise the report. Its main goal was to complete the report and tease out conclusions and recommendations for future research in this area. The continuation of the work of the Task Force was also discussed.

1.6. Outline of the report

The present report summarises the work carried out by the “Radioecology and Waste” Task Force in the scientific workshops (members of the Task Force and participants in the workshops are listed in Appendix 3). In Section 2 an overview of the behaviour of the studied radionuclides is presented with the help of interaction matrices that were developed by the group for terrestrial environments (Appendix 1) and aquatic environments (Appendix 2), respectively. The suggestions of the group as to general approaches for improvement of the assessments models are summarised in Section 3, while the needs for research and development identified by the group for each specific radionuclide are presented in Section 4. The main conclusions and recommendations of the work are given in Section 5.

2. Overview of the Environmental Behaviour of ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U

Interaction matrices (Hudson, 1992) were used during the scientific workshops to facilitate the discussions and as a way to present the information in a synthesised and structured way. Matrices for terrestrial and aquatic environments were produced to identify relevant processes and rank these in order of significance. The ranking of processes by their significance proved to be a difficult process, mainly because the criteria and scope of the ranking were not clearly stated. The group decided to use a simple classification, where the processes were divided into two categories: potentially important and unimportant. The group analysed in more detail the processes classified as potentially important. The final aim was to identify ways of improving assessment models on the basis of a better understanding gained through experimental research.

In developing an interaction matrix, the first step is to identify the main components (or features) of the system to be analysed. These components comprise the leading diagonal elements (LDE) of the interaction matrix. The off-diagonal elements (ODE) are used to describe the processes that relate the main components. The matrices should be read clock-wise.

2.1. Interaction matrix for terrestrial environments

Figure 2.1 shows the general matrix representation of the migration of radionuclides in a terrestrial ecosystem. Appendix 1 illustrates terrestrial matrices for individual radionuclides (^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U).

The major components of the terrestrial environment are closely related to those discussed in BIOMASS (2003), but plants and animals are treated as distinct components rather than being aggregated as biota and soils are disaggregated into several distinct components. The leading diagonal elements in the matrix are described below.

- The **atmosphere** comprises the region above the soil surface. It includes both the region below the plant canopy and the region above it.
- **Water bodies** include surface water bodies, e.g. rivers, streams and lakes, and subsurface water bodies such as aquifers.
- **Vegetation** comprises macroscopic vegetation.
- **Animals** comprise those living in the terrestrial environment both on the soil and burrowing within it. Thus, this category includes farm animals (e.g. cattle and sheep), domestic animals (e.g. dogs), herded animals (e.g. reindeer), wild animals (e.g. foxes) and burrowing biota (e.g. rabbits, moles and earthworms).
- **Soil** is defined as the unconsolidated mineral or organic material on the immediate surface of the Earth that serves as a natural medium for the growth of land plants[1].

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[1] Soils are considered as living systems (in contrast to the historical perspective of soil as inert mineral material); include complex assemblage of organisms and by-products. Soils are dynamic, continually aggrading and degrading. Some changes occur over very long time scales (weathering of primary minerals, formation of secondary minerals), and others occur on very short time scales (root and microbial uptake of inorganic nutrients). Physical structure is an important aspect of soils. At a coarse scale this includes soil profile structure (kinds and depths of various horizons), and at smaller scales it includes soil texture, aggregate size distribution and stability, porosity, etc.

- **Soil Solution** comprises the aqueous medium present in pores in soil. It can be derived from meteoric water or from groundwater. Its composition is conditioned both by its source and by the materials and biota with which it is in contact.
- **Soil Atmosphere**. The content of air in a soil sample. In a well-aerated soil, the soil air is very similar in composition to the atmosphere above the soil. Poorly aerated soils usually contain a much higher content of CO_2 and a lower content of O_2 than the atmosphere above the soil. The rate of aeration depends largely on the volume and continuity of air-filled pores within the soil.
- **Soil Organic matter.** The organic fraction of the soil exclusive of undecayed plant and animal residues.
- **Soil microbiota** comprises microflora and protozoa, but not larger organisms such as earthworms and burrowing mammals.
- **Soil inorganic matter** comprises soil minerals. Each soil mineral is defined as a natural inorganic compound with definite physical, chemical and crystalline properties (within the limits of isomorphism).
- **Interface with the geosphere.** This is an artificial frontier[2]. In terrestrial environments, it will be normally represented by weathered saturated or unsaturated materials.

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[2] See Section 3.5

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)
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
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) Eructation	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)
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)
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
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
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
1)Resuspension (at outcrop)	1)Desorption 2)Mineral dissolution	1)External contamination	1)Ingestion 2)Bioturbation	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

Figure 2.1: General matrix representation of the migration of radionuclides in a terrestrial ecosystem.

2.1.1. Processes in the ODE of the Terrestrial Matrix and their definition

The various processes in the off-diagonal elements of the interaction matrix, shown in Figure 2.1, are classified in Table 2.1 as being physical transport, a chemical interaction, a biological process or an intake route.

Table 2.1: Transfer processes included in the ODE of the terrestrial interaction matrix.

Physical transport		Chemical interactions	Biological processes	Intake routes
Advection Capillary rise Colloid transport Death Degassing Deposition (dry and wet) Diffusion Diffusive exchange Droplet production Eructation Evaporation Excretion Exhalation External contamination Gas evolution Gas sorption Groundwater recharge	Irrigation Leaching Leaf fall Mechanical weathering Particle transport Pollen and seed release Pressure pumping Recharge by surface water Release due degradation Release from solution Release of OM Respiration Resuspension Seepage Solution at boundaries Through-flow Transpiration	Adsorption Carbonate production Chemical weathering Complex formation Desorption Fermentation Fixation Ion exchange Isotopic exchange Mineral dissolution Mineral precipitation (mineralisation) Precipitation Solution Sorption	Biofilms Biological weathering Bioturbation Excretion of microbiota Fertilisation Microbial metabolism Photosynthesis Root exudation Root respiration Root uptake Symbiotic association Transport by arenchyma Transport by microbiota Utilisation	Inhalation Ingestion Uptake

The transfer processes can be defined as follows:

- ***Adsorption***. 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 or soil atmosphere and retained on the surfaces of solids by chemical or physical binding. Radionuclides present in the soil atmosphere can potentially adsorb directly onto organic matter. However, adsorption will typically occur following transfer from soil atmosphere to soil solution.
- ***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 aquifer.

- ***Biofilms***. Microbial populations generally live within biogenic polysaccharide films (biofilms) on surfaces. A biofilm will only exist as long as there are sufficient nutrients and energy sources and available water. Biofilms can vary in depth (from one cell thickness to several cm) depending on the environment and will contain biocommunities. The physico-chemical environment changes within the biofilm enable a variety of microbial species to live together, often with each species being dependent on another. Given sufficient nutrients, energy sources and available water, biofilms will form on surfaces. The biocommunity present will affect the water chemistry, influence mineral dissolution and mineral precipitation, and generate inorganic colloids with resulting influences on radionuclide speciation. Biofilms will also have physical impacts e.g. physical retardation, which will influence water movement in the soil system.
- ***Biological weathering*** can be caused by macro and micro organisms. On the macro scale, plant roots can penetrate cracks and fissures in rocks, forcing them apart. On the micro scale, rock surfaces are often colonised by a biocommunity of algae, fungi, lichens (algae and fungi living together) and bacteria, which often live together in a biofilm.
- ***Bioturbation***. The redistribution and mixing of soil or sediments by the activities of plants and burrowing animals. All living organisms can bioturbate their environment as a result of their normal biochemistry and life processes. Bioturbation effects are, in general, likely to increase the transport of radionuclides.
- ***Capillary rise***. Upward movement of water through soil layers above the water table as a result of capillary forces related to evaporation and transpiration. This is a water transport process that is the result of water adhesion and surface tension in a porous medium. Water adhering to the walls of a soil pore will cause an upward force (surface tension) on the liquid at the edges. Capillary action occurs when the surface tension is stronger than the cohesive forces between the liquid molecules. A capillary fringe is the soil area just above the water table where water can rise up slightly through capillary action. This layer ranges in depth from a few centimetres to a couple of metres, and it depends on the pore sizes of the soils/rocks involved. Fine-grained materials produce larger capillary action effects. The rise of water in a capillary fringe may cause the upward migration of dissolved radionuclide solutes and possible mineral precipitation or dissolution at the edge of the capillary fringe.
- ***Carbonate production***. ^{14}C may be incorporated directly in inorganic soil solids by precipitation as carbonate.
- ***Chemical weathering***. The breakdown of rocks and minerals due to the presence of water and other components in the soil solution or changes in redox potential
- ***Colloid transport***. The complexation of materials can form colloids. Biocolloids are stable dispersions of biological particles in groundwater. They can be produced by a number of microbial metabolic processes e.g. by generation of organic by-products. Microbes themselves can also act as biocolloids and can be transported passively and/or actively (where they can control their own movement e.g. towards a nutrient source). Some biocolloids may be introduced to the Geosphere-Biosphere Interface (GBIZ) with groundwater from the biosphere. Consequently, biocolloid populations will be higher in the near-surface ground and in pore waters than in deeper groundwaters from more stable environments. Radionuclides can adsorb onto biocolloids in a similar way to inorganic colloids. Thus, if a radionuclide is poorly soluble but is adsorbed onto a mobile biocolloid, its transport will be greatly enhanced. However, most microbes tend to attach to surfaces in biofilms thus, in this mode; a biocolloid will tend to decrease radionuclide transport.
- ***Complex formation***. A complex is a combination of two substances, which normally remains dissolved.

- ***Death***. Animals that die release radionuclides to their immediate environment after decomposition.
- ***Degassing*** (out-gassing). Soil gas emission. Out-gassing is the mechanism by which gases are released from the Earth's surface into the atmosphere. Out-gassing may lead to bubble-mediated transport or gas-driven groundwater flow. Out-gassing potentially increases radionuclide transport rates to the surface. Degradation of organic matter can lead to gas production and releases of gases containing radionuclides to the soil atmosphere. Radionuclides such as ^{14}C and ^{79}Se have the potential to be involved.
- ***Deposition***. The removal of gaseous or particulate material from the atmosphere by precipitation, causing deposition of material onto surfaces. Radionuclides can be deposited directly from the atmosphere as a result of dry deposition of particles or gases and vapours, or by wet deposition in which the radionuclide is either captured 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 significant 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 cause 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.
- ***Diffusive exchange***. Radionuclides present in gas or vapour form in the above-ground atmosphere can exchange with the soil atmosphere by diffusion.
- ***Droplet production***. Radionuclides can be released from the surfaces of water bodies such as rivers and lakes by the creation of droplets either from bubble bursting or from waves.
- ***Eructation***. Release of radioactive gases produced in the gastrointestinal tract (notably ^{14}C-methane in ruminants) during digestion.
- ***Evaporation***. Transfer of water from the ground directly to the atmosphere. Tritium in soil solution could be released to atmosphere directly by evaporation. Other radionuclides might be transferred to the gas phase in soil atmosphere and then released. However, this is an indirect route via soil atmosphere. Radionuclides can be released from the surfaces of water bodies such as rivers and lakes by evaporation.
- ***Excretion***. Although most of the material excreted by animals is organic in nature, inorganic materials such as soil particles are ingested, pass through the gastrointestinal tract and are eventually excreted[3].

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[3] Excretion of gut microbiota. Animals contain an active microbiota in their gastrointestinal tract. Uptake of radionuclides can occur into this microbiotic community and some of the microbiota can be excreted. Urinary excretion can directly contaminate soil solution, as can the liquid component of faecal excretion. Faecal excretion will result in direct entry of radionuclides incorporated in organic solids into soil. Similarly, the death and decomposition of animals results in the incorporation of organic solids in soils. These comments apply both to animals present on soils and to the soil macrofauna (e.g. burrowing mammals and earthworms) present within soils.

- ***Exhalation***. Animals may release radionuclides to atmosphere by immediate exhalation of inhaled activity that is not deposited in the respiratory system, by exhalation of radioactive gases produced by metabolic processes in tissues.
- ***External contamination***. Solid organic matter from soils may be present on the external surfaces of vegetation.
- ***Fermentation***. See Respiration.
- ***Fertilisation***. The import of artificial fertiliser to enhance crop productivity.
- ***Fixation***. Radionuclides present in soil solution may be irreversibly incorporated in soil inorganic matter by chemical or biological processes.
- ***Gas[4] evolution***. Radionuclides can be released from the surfaces of water bodies such as rivers and lakes by gas production within the water body and release from it (e.g. ^{14}C).
- ***Gas sorption***. Radionuclides in gas or vapour form may be taken up directly into soil solution present at the soil surface, though it would be more usual for such uptake to be indirect via the soil atmosphere.
- ***Groundwater recharge***. The percolation of incident precipitation and other surface waters to groundwater systems.
- ***Ingestion***. Incorporation of radioactivity into the body in water or other contaminated substances by ingestion. Animals may deliberately or adventitiously ingest soil and, therefore, soil microbiota, soil solution and soil inorganic matter. Herbivores and omnivores ingest living vegetation. Detrivores ingest senescent vegetation. Animals may ingest radionuclides present in drinking water also.
- ***Inhalation*** (burrowing animals). Incorporation of radioactivity into the body in the form of aerosols, vapours or gases as a result of breathing. High concentrations of radionuclides in air may build up in the excavations of soil-inhabiting macrofauna. These high concentrations may be inhaled. ^{222}Rn and its progeny originating from ^{226}Ra present in soil may be of particular interest in this context.
- ***Inhalation by animals***. Animals can inhale radionuclides present in particulate form (as micron or sub-micron size aerosols) or as gases and vapours.
- ***Ion exchange***. Ion exchange may be considered a special case of sorption. It occurs when an aqueous radionuclide ion displaces a counter-ion in the substrate surface layer and is retarded. This process is especially important for positively charged radionuclide species interacting with clay minerals that have a high cation exchange capacity (CEC).
- ***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 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.
- ***Isotopic exchange and solution***. Radionuclides may move from the soil atmosphere to soil solution either by isotopic exchange with other atoms of the same element or by dissolving in the soil solution.
- ***Leaching***. The removal of soluble materials from one zone in soil to another via water movement in the profile.

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[4] Gas transport processes include diffusion, convection, two-phase flow and bubble formation. The partial pressure gradient, molecular weight and temperature, control the rate of migration of a gaseous species by gas diffusion. The pressure differential is the overall factor governing gas flow by convection. Gas transport usually relies on unsaturated media to be present, as water offers an effective barrier to gaseous transport. In saturated media, gas transport may occur through the formation of bubbles.

- ***Litter fall.*** During senescence and death, plant material enters soil. This is often initially into the litter layer. However, the material then decomposes and bioturbation processes mix it into soil organic matter. This material may be rapidly degraded in soil, or it may be either physically or chemically stabilised and only slowly degrade.
- ***Mechanical weathering.*** Mechanical disintegration of a material. Physical weathering may be a precondition of any chemical weathering of a rock. The physical agents that can cause mechanical disintegration of an exposed material are temperature fluctuations, wind, water and ice flow. Diurnal and seasonal temperature changes may cause repeated expansion and contraction leading to exfoliation or spalling of a rock surface. If a material is wet, then a freeze-thaw process (frost shatter and frost wedging) may disrupt its physical integrity. Freeze-thaw processes depend on ice expanding as the water freezes in the joint or fracture. The process of mechanical weathering breaks down a rock into smaller fragments; no chemical change is involved.
- ***Microbial metabolism*** may convert radionuclides in soil inorganic matter to being in organic matter. However, this would usually involve the intermediate step of incorporation of the radionuclides in the microbial biomass, so this is probably best represented as an indirect pathway. Metabolism is a basic characteristic of all living systems; metabolic reactions (particularly those producing energy) keep the cell alive. Metabolic reactions occur in small steps (metabolic pathways) so that the organism can derive as much benefit from each reaction as possible. Many of these reactions are catalysed by enzymes. Two types of reactions occur in living systems: synthetic and breakdown. Synthetic reactions comprise anabolism and breakdown reactions those of catabolism. Anabolic reactions require energy (and build up structures etc within an organism) whereas catabolic reactions produce it (to drive anabolic reactions, to provide energy for work and for maintenance of the organism).
- ***Mineral Dissolution and Co-dissolution.*** Process by which material in the solid phases is incorporated into the liquid phase. Affected by local Eh, pH, solubility limits and the presence of other chemical species. 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 stochiometry 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.
- ***Particle transport.*** Natural transport processes causing movements of solid material between environmental media.

- ***Photosynthesis***. If ^{14}C is present in the atmosphere it can be incorporated directly into plants as a result of photosynthesis. Also, other gases and vapours, such as sulphur dioxide, can penetrate directly into plants through the stomata and be taken up by the leaf mesenchyme. This could be relevant to the deposition and uptake of radionuclides such as ^{35}S and ^{79}Se.
- ***Pollen and seed release***. Radionuclides incorporated in solid parts of plants can be transferred to atmosphere with those plant parts. In some cases, it may be convenient to treat such transfers as directly from plants to organic matter in soils (see release of organic matter). However, some plant parts such as pollen, seeds and leaves may be transported considerable distances through the atmosphere before deposition.
- ***Precipitation***. Rain, snow, hail etc. as part of the natural hydrological cycle. Radionuclides may be dissolved in wet precipitation and, therefore, enter soil solution directly.
- ***Pressure pumping***. Radionuclides present in gas or vapour form in the above-ground atmosphere can exchange with the soil atmosphere by advective pumping induced by changes in air pressure forcing gas into or out of the soil zone.
- ***Recharge by surface waters***. The percolation of incident precipitation and other surface waters to groundwater systems. Surface waters may flow across the soil surface, e.g. as a result of infiltration excess or flooding. Such waters could contain radionuclides in solution (or attached to suspended solids) that could be transferred to soil solution. Surface waters could flow over outcrops of bedrock. If the outcrops were unsaturated at the surface, recharge would occur carrying radionuclides into the bedrock. The rate of groundwater recharge depends on the type of subsurface rock materials in a given area. Saturated permeable layers capable of providing a usable supply of water are known as aquifers. Typically, they consist of sands, gravels, limestones, or basalts. Layers that tend to slow down groundwater flow, such as clays, shales, glacial tills, and silts, are instead called aquitards. Impermeable rocks are known as aquifuges, or basement rocks. In permeable zones, the upper surface of the zone of water saturation is called the water table Recharge is from atmospheric precipitation and percolation of the water through the overlying soil and rock sequence. The rate of groundwater recharge depends on the type of subsurface rock materials in a given area.
- ***Release during degradation***. Decay of the organic matter may result in radionuclide releases that cannot be characterised as reversible.
- ***Release from solution***. Water bodies present in contact with soil could degas at their boundaries, releasing radionuclides into the soil atmosphere. However, it would be more usual for water to enter soil, so becoming part of soil solution, and then degas.
- ***Release of Organic Matter***. Organic matter may be carried downward in percolating water. This may be as particles or as colloidal humic materials.
- ***Respiration and fermentation processes***. Radionuclides may be released from soil microbial communities in gaseous form. This is potentially important for ^{3}H and ^{14}C (e.g. in methane and carbon dioxide), but may also be of significance for other radionuclides, e.g. ^{36}Cl, ^{79}Se and ^{129}I. Gases can be formed by microbial activity in three ways: direct biodegradation of organic materials (which act as a nutrient source); direct catalysis of anaerobic corrosion of metals; and indirectly by producing chemical environments which cause gas production e.g. production of acids which enhance metal corrosion. Carbon dioxide, hydrogen sulphide and methane are examples of biogases directly produced by a variety of different microbial species living together in a biocommunity within a biofilm.
- ***Resuspension***. Solid organic matter, microbial community and solid inorganic matter in soils may be resuspended to atmosphere by the wind and human activities. Parent material may also be resuspended to atmosphere. This could occur where it is exposed

at outcrop and has been weathered to a fine enough consistency to make it susceptible to resuspension.

- ***Root exudation***. Roots may penetrate into parent material, so exudation can release organic compounds and radionuclides to this zone. In addition, plants can develop in pockets of soil on outcrops and can contribute biotic material on senescence and death. Vegetation with its roots penetrating surface water bodies or deep groundwater may transfer radionuclides by exudation into the water body. Organic compounds produced by roots are released into the soil. These compounds may incorporate radionuclides. However, in addition, such compounds may play a role in mobilising radionuclides present in the rhizosphere and making them more available for plant uptake.
- ***Root respiration*** can release ^{14}C-labelled carbon dioxide into the soil atmosphere. However, other volatile gases that are products of metabolism and are labelled with radionuclides such as ^{35}S and ^{79}Se may also be released.
- ***Root uptake***. 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. Some plants have root systems that penetrate directly into surface water bodies. These roots may take up radionuclides directly from those water bodies. In addition, deep rooting plants may have roots that penetrate to groundwater within parent material, taking up radionuclides from that source.
- ***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***. Vegetation with its roots penetrating surface water bodies or deep groundwater may transfer radionuclides by death of the plant or senescence of parts leading to releases into the water body. This includes falls of branches. However, deposition of leaves, seeds and pollen onto water bodies is best considered as occurring via the atmosphere.
- ***Solution at boundaries***. Radionuclides present in the soil atmosphere may diffuse into surface water bodies across the soil boundaries and go into solution.
- ***Solution***. See isotopic exchange.
- ***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.
- ***Symbiotic association***. The boundary between plant roots and soil microbiota is not well-defined. Mycorrhiza form a symbiotic association between fungi and plant roots, so nutrients and radionuclides may be exchanged directly between plants and the soil microbiota by this route.
- ***Through-flow***. 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.
- ***Transpiration and Respiration***. Transpiration is the transfer of water from the soil to the atmosphere by plants. Vegetation can release radionuclides to atmosphere in the transpiration stream (^{3}H) or by respiration (^{14}C as carbon dioxide and other radionuclides such as ^{35}S and ^{79}Se incorporated in gaseous products of metabolism).
- ***Transport by microbiota***. May move downward to invade parent materials as part of the process of soil formation. Migration may be by active or passive transport. Such

migration can carry radionuclides with it. In addition, microbial populations present in parent materials may increase by reproduction.

- ***Transport in aerenchyma***. Plants can contain inter-connected gas-filled pathways (aerenchyma), particularly in water-logged soils. These are a potential route of transport for radionuclides from the soil atmosphere to plant tissues.
- ***Uptake***. Nutrients and radionuclides in the soil atmosphere may be directly available for uptake and incorporation in soil microbiota, but this is likely to be secondary compared with uptake from soil solution. Soil solution constitutes a primary source of nutrients for soil microbiota. Thus, radionuclides in soil solution may be highly available for incorporation in microbial biomass.
- ***Utilisation***. Soil organic matter may be directly utilised as a substrate by soil microbiota. Thus, radionuclides may pass directly from organic matter to microbiota without passing through soil solution.

2.2. Important processes in terrestrial environments

This section provides an overview on the processes that were identified as potentially important for the long-term behaviour of the studied radionuclides (^{14}C, ^{36}Cl, ^{237}Np and ^{238}U) in terrestrial environments. The general interaction matrix for terrestrial environments, Figure 2.1, was used for identifying potentially important processes for individual radionuclides, shown in Appendix 1. Below the main findings from the discussion during the scientific workshops are summarised.

2.2.1. ^{14}C

The dynamics of carbon behaviour in terrestrial ecosystems are well understood. The following processes were considered to have the highest importance for describing the cycling of ^{14}C in terrestrial ecosystems: photosynthesis, respiration by the above-ground vegetation and animals and the soil biota, fermentation by animals and soil biota, exhalation and diffusion of gases from the soil atmosphere, ingestion of vegetation by animals, litter fall and senescence, ingestion and utilisation of soil organic matter by soil biota.

Post-closure radiological assessments of solid radioactive waste disposal have shown that the generation of ^{14}C-methane and ^{14}C-carbon dioxide can be of radiological significance. These gases are produced by microbial degradation of wastes and can be transported to the surface together with hydrogen produced from metal corrosion in reducing conditions. If continuous gas-filled pathways are produced, such transport can be rapid. ^{14}C-carbon dioxide may be a limited problem, as it would be expected to react with a cement/concrete in the near field. However, ^{14}C-methane is likely to be non-reactive in both the near field and geosphere, but will be subject to microbial metabolism to ^{14}C-carbon dioxide in the soil zone. In addition, ^{14}C may be released from a repository in solution in groundwater. Such groundwater may be extracted from wells and used for irrigation and other purposes. It is not clear how much of the ^{14}C would be lost from extracted groundwater before that groundwater was utilised. In particular, losses from solution during spray irrigation might be substantial.

Although the majority of plant carbon is generally obtained from the atmosphere by photosynthesis, significant root uptake has been observed and this may contribute up to a few percent of plant carbon.[5] Also, if ^{14}C-carbon dioxide is released from soil, its

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[5] As early as 1954, Livingston and Bell (1954) stated that: “The conclusion emerges that ordinary green plants with roots in soil or solution and leaves or other transpiring organs exposed to the air probably do not usually derive from the free air all of the carbon dioxide required by photosynthesis, but that a portion of that carbon dioxide is generally derived from the soil and, to some extent, from the plant’s own respiration; also that the portion of carbon dioxide not derived from the free air may amount, in some instances, to at least 5% of the total amount”.

concentration in the entrapped sub-canopy atmosphere may be much higher than in the above-canopy atmosphere.

Some ^{14}C entering the gastrointestinal tract may be completely broken down on ingestion and enter the bicarbonate pool in tissues. However, a substantial proportion may bypass the bicarbonate pool and be incorporated directly into carbon compounds in tissues. Loss from the bicarbonate pool occurs rapidly to exhaled air. Thus, although ^{14}C is generally completely taken up from the gastrointestinal tract, considerable distinctions exist relating to its subsequent partitioning between different forms and hence in its uptake and retention in tissues.

The classical approach for estimation of the halftime of ^{14}C in farm and wild terrestrial animals, based on body carbon content and digestible carbon intake, is an underestimation of real organic carbon metabolism by a factor up to 2. For farm and wild animals the whole body carbon halftime can be expressed using mass allometric relationships. For farm animals it is important to consider the effect of maturity on the carbon half times.

There exist good models of carbon cycling in all types of ecosystem, which could be adapted to the problem of long-term ^{14}C cycling. The selection of the appropriate model is a matter of defining appropriate time and spatial scale for the problem. For long time scales, consideration of the effects of climate change will be necessary, especially if there is an interest in protection of the environment.

[5 (cont'd)] On the basis of studies in established sunflower plants, Sauerbeck and Fuhr (1966) estimated that during the course of a 6 day experiment approximately 1.3 % of overall carbon assimilation occurred through the roots.

Reports from the Macaulay Institute (1980) suggest that it is possible that up to 10 % of the ^{14}C content of ryegrass grown in soil amended with labelled polymaleic acid arises from labelled carbon dioxide in the soil matrix derived from decomposition of the polymer.

Morgan and Beetham (1990) reported also on work in their own organisation (Associated Nuclear Services, ANS) and gave an interpretation of all the work that they had reviewed. The relevant text from their report is reproduced below.

Over the period 1984-1988, ANS investigated the uptake of ^{14}C by ryegrass from both solution and soil using carefully-controlled systems to ensure that labelled ^{14}C could enter plants only by root absorption from contaminated soil rather than by foliar uptake of $[^{14}C]O_2$ released from soil (Kirton *et al.*, 1989). These studies provided conclusive evidence for the uptake and translocation of ^{14}C from the soil and solution culture bicarbonate pools over relatively short timescales. It is probable that total uptake did not amount to more than a few percent of that present in soil or solution, though further experimental work is required to confirm this.

It is clear from the above [review] that there is ample evidence to demonstrate that ^{14}C present as bicarbonate or carbon dioxide in soil can be absorbed by plant roots. According to growth conditions, a substantial fraction can be translocated to shoots, possibly *via* synthesis into malic acid. The underlying mechanisms and processes of this uptake (particularly the role of the rhizosphere) and translocation have not been investigated in detail, despite the obvious implications for plant growth and agriculture (e.g. Livingston and Bell, 1954; Berquist, 1964). There appear to be species differences in the ability to absorb and translocate carbon from the soil carbonate pool. One possible factor of significance is the degree to which air spaces and aerenchyma tissues are developed.

Overall, it seems reasonable to estimate that about 1 to 10 % of plant carbon may derive from the soil. With a biomass production of 1-5 kg (fresh weight) m^{-2} y^{-1}, this would represent removal of 0.0004-0.02 kg (C) m^{-2} y^{-1}. For comparison, Smith and Smith (2001) give the detrital carbon content of temperate grasslands as 19.2 kg (C) m^{-2}. Thus, the total uptake by plants from soil in such systems corresponds to no more than 0.1 % of the content per year. For mineral soils, about 2.4 kg (C) m^{-2} is a more reasonable estimate of the carbon inventory, implying an uptake of up to about 1 % of the content per year.

The use of concentration factors in its classical definition to describe ^{14}C transfer is not recommended. Instead alternative approached, such as the specific activity approach and dynamic models should be given preference. This is discussed in more detail in Section 3.

2.2.2. ^{36}Cl

As reflected in the corresponding matrix, see Appendix 1, the processes leading to vertical redistribution in the system were considered key processes for the long-term behaviour of ^{36}Cl in terrestrial ecosystems. This concerns the parameters influencing the recycling of ^{36}Cl in the soil-subsoil system, the balance of the inputs and outputs of stable chlorine in the system and the remobilisation (leaching) of chlorine from soul organic matter due to decomposition.

Two recent experimental studies that were carried out as part of the MITRIC-UA project “Studies of transfer of halogens to plants”, financed by IRSN, have assessed and improved knowledge of the behaviour of ^{36}Cl in terrestrial ecosystems:

- A study that focused on ^{36}Cl behaviour in the soil-plant system, with especial emphasis on the following issues: (i) the absence of sorption in soil, ii) a prevailing role of fixation by biota and iii) the migration with the soil moisture flux, (iv) the root uptake by plants, which is characterised by quite high concentration ratios, v) the dependence of ^{36}Cl behaviour in the environment on stable chlorine content.
- A study of the upward soil migration and plant uptake of ^{36}Cl, which showed that, compared to a fixed water table, the increase of the water table height leads to substantial increase of the water fluxes, and hence ^{36}Cl migration, up through the soil. Estimates of the distribution coefficients (K_d values) in soils yielded values close to zero. Over time, the soil profile of ^{36}Cl tended towards a uniform vertical distribution. Once present within the rooting zone of the soil column, ^{36}Cl was readily taken up by perennial ryegrass growing on the surface of the column. Concentration ratios from soil to plants (expressed in Bq.kg^{-1}(dw).in plant per Bq.kg^{-1}(dw) in soil) of around 200-400 were measured.

Extensive studies have been undertaken on the natural origin of organic chlorine compounds (see, for example, Öberg, 2003; Öberg *et al.*, 2005). Therefore, incorporation of ^{36}Cl by soil biota in organic matter should be considered to fully understand the behaviour of ^{36}Cl in soils.

The balance of the input and output of stable chlorine in the system is an important factor that needs to be taken into account when modelling ^{36}Cl migration in the environment. It is also important to improve the description of the dynamics of chlorine remobilisation (leaching) from soil organic matter due to disintegration the organic forms.

2.2.3. ^{99}Tc

The behaviour of Tc in the environment is dictated by its chemical speciation. There are few data available on K_d values in different redox conditions. The data of soil-to-plant concentration factors is also limited, especially for non-agronomic, natural, plants. The behaviour of Tc in humic systems has been poorly studied.

Results of an experimental study, conducted at Imperial College, on the upward soil migration and uptake of technetium from contaminated groundwater, have shown that chemical reduction of Tc(VII) leads to the precipitation of insoluble Tc(IV) species and, therefore, there is limited soil migration from groundwater and through an anoxic/oxic front. However, slow upward migration of the reduced species seems to occur, which could be expedited by soil re-oxidation (receding water table) or soil management (drain installation, ploughing). The observed plant uptake is low due to the low degree of migration – plant roots seemed not to be reaching the contaminated soil depths. These results are in contrast to the generally held view that Tc is very soluble and mobile in soils.

2.2.4. ^{237}Np

There are few environmental data for neptunium; due to difficulties of measurement in the past. There are thus considerable gaps in the understanding of the environmental behaviour of the element. This is reflected in the interaction matrix for this radionuclide, Appendix 1, where there is no clear differentiation in the importance of the transfer processes.

In some conditions, Np seems to have higher mobility than other actinidies, like Pu and Am. Also, it is possible that enrichment of Np takes place in specific environments, as it is the case for other actinides, especially those that are redox-sensitive, e.g. Pu. In the context of HLW disposal, such environments are of particular interest.

With current analytical techniques, it should be possible to perform studies of the Np behaviour in foodchains, which are missing at the moment. The deposition of Np from Nuclear tests fall-out and Chernobyl has been investigated by using lichens. It would be a natural step to go further to different organs of reindeer. By doing that, it would be possible to assess the gastrointestinal uptake and estimate residence times under natural conditions. Our present knowledge of Np behaviour in the human body is based mainly on experimental studies with rats. A study of reindeer is not ideal to describe the situation for man, but it would extend the range of information available. There are short-lived Np isotopes that could be used with minimal doses to study uptake and retention in man. There are also autopsy samples from the early fallout periods that could be used.

The lack of currently available environmental data on neptunium is emphasised. The low levels of this radionuclide in the environment mean that studies have largely been limited to the plume of Np from Sellafield. All the studies done so-far point to the fact that neptunium may be more soluble than Am and Pu in oxidising systems, but that when reduction occurs, Np solubility may decrease. This is particularly important to consider in the context of waste management scenarios.

2.2.5. ^{238}U

Soil variables and the plant age and species seem to be important determinants of uranium uptake and further distribution within the plant (Shahandeh *et al.*, 2001). The levels of carbonate, phosphate, organic matter and oxy-hydroxides of Fe and Al in soils greatly influence uranium solubility and mobility. U forms highly soluble complexes with carbonate in calcareous soils, inducing a higher U availability for plants compared with more acidic soils devoid of carbonates. Conversely, P fertilisation of U-contaminated soils has been shown to induce a significant decrease of the water-soluble and extractable U with, as a result, a reduction of the net uptake by plants while the biomass production is improved (Rufyikiri *et al.*, 2006). Uranium uptake and possible consequent toxic effects are probably better related to soluble and easily exchangeable level in soils rather than total uranium. In hydroponics, U quickly becomes associated with roots. Some of the uranium is absorbed and translocated within the plant, but usually a dominant portion remains at the surface of, or in, the roots. Graminoid (grass-like) plants can show significant uptake in the above-ground parts whereas woody plants showed exceedingly high uranium accumulation in the root portions. In trees, uranium seems to be translocated as a strong soluble complex in xylem sap and mainly accumulates in foliage without particular dilution processes or concentration episodes resulting from internal translocation. The transpiration stream is most probably the determinant factor responsible for increasing U content corresponding with the age of the evaporative organs. The senescing foliage was shown to act as a dominant sink for the U moved from the roots. As a consequence, litterfall plays a major role in tree detoxification and is a major pathway in the recycling of U to the soil (Thiry *at al.*, 2005). The formation of stable complexes or precipitates is likely the main mechanism of uranium accumulation in roots as well as in other plant parts and in rhizospheric fungal microbiota, and this is assumed to contribute to the low translocation of uranium. Many studies have shown that U sorption by fungal mycelia or bacteria can be important and then greatly influence the availability of U in soil and in the plant rhizosphere. An adequate mycorrhization of crop plants generally decreases U translocation from root to shoot and can thus be of complementary use in the revegetation of U-contaminated sites. The sequestration of U in intraradical fungal hyphae was also evoked as a mechanism of arbuscular mycorrhizal protection against U toxicity for plants (Rufyikiri *et al.*, 2005).

The empirically derived values of K_d and the soil-to-plant TF of uranium show an extremely high variability and are strongly dependent on soil pH. A satisfactory explanation of this variability has not yet been found. In particular, the role of soil-plant-microbiota interactions and the physiological parameters controlling uranium translocation in plants are poorly understood.

There is a large amount of useful data for uranium, such as data of K_d and CR, covering a wide range of environmental conditions. The variability in the data is, however, very large, reflecting the large variability observed in the speciation of uranium. Attempts to reduce the variability have been made, for example by dividing the K_d values into different categories according to the pH of the media (see Section 3.1). Further refinements of such categorisation might be achieved by considering dependencies on multiple variables. A similar categorisation might be done for the CR to biota, but it will be necessary to adopt more mechanistic experimental and modelling approaches (Sheppard *et al.*, 2006). It could be opportune to distinguish between the pool of U adsorbed on the roots from the pool inside root tissues and from that translocated to above-ground plant parts, as it seems important to differentiate between the uranium that is on the surface and inside the biota, for instance the fraction present on the surface of the plants due to soil resuspension and inside the plants due to root uptake.

For long-term assessments it is also necessary to take into account the issue of uranium equilibration with its daughters. The pathways associated with the daughters will have to be considered and the models appropriately parameterised.

2.3. Interaction matrix for aquatic environments

Figure 2.2 describes the general matrix representation of the migration of radionuclides in aquatic ecosystems. Appendix 2 illustrates aquatic matrices for individual radionuclides (^{14}C, ^{237}Np, ^{36}Cl, ^{99}Tc and ^{238}U).

Leading diagonal elements in the matrix are described below.

- The A**tmosphere** comprises the region above the soil surface.
- **Water** includes surface water bodies, e.g. rivers, streams and lakes.
- **Abiotic Suspended Matter** are inorganic particles > 0.45μm suspended in water.
- **Deposited Matter (sediment)** is particulate matter (inorganic and organic) deposited in the bottom of rivers, streams and lakes, including interstitial water.
- **Primary Producers** (phytoplankton, macrophytes, and aquatic plants) are autotrophic organisms.
- **Consumers I** (zooplankton, macrobenthos) are animals that feed on Primary producers.
- **Consumers II** (omnivorous fish) are animals that feed on Consumers I.
- **Decomposers** comprise microflora and protozoa, but not larger organisms such as benthic invertebrates.

Atmosphere	1)Deposition							1)Diffusive exchange 2)Pressure pumping
1)Evaporation 2)Gas evolution 3)Droplet production	Water	Adsorption, complexation, precipitation	1)Diffusion 2)Advection 3)Adsorption 4)Complexation 5)Precipitation	1)Uptake 2)photosynthesis 3)deposition	1)Uptake	1)Uptake	1)Uptake	1)Recharge by surface waters
1)Resuspension	1)Desorption 2)Dissolution	Abiotic Suspended Matter	1)Deposition		1)Ingestion	1)Ingestion	1)Ingestion 2)Uptake	1)Particle transport 2) Colloidal transport
1)Resuspension	1)Desorption 2)Dissolution 3)Degassing	1)Erosion of bed sediment	Deposited Matter (sediment)	1)Root uptake	1)Ingestion	1)Ingestion	1)Ingestion 2)Uptake	1)Particle transport 2) Colloidal transport
1)Respiration 2)Release of other organic matter	1)Respiration 2)Release of other organic matter		1)Death	Primary Producers (phytoplankton, macrophytes, aquatic plants)	1)Ingestion	1)Ingestion	1)Ingestion 2)Uptake	1)Biological weathering
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		1)Death 2)Bioturbation		Consumers I (zooplankton, macrobenthos)	1)Ingestion	1)Ingestion 2)Uptake	1)Excretion 2)Death and decomposition
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		1)Death 2)Bioturbation			Consumers II (omnivorous fish)	1)Ingestion 2)Uptake	1)Excretion 2)Death and decomposition
1)Resuspension	1)Resuspension 2)Excretion		1)Death 2)Bioturbation		Grazing/Uptake		Decomposers	1)Excretion 2)Death and decomposition
1)Resuspension	1)Desorption 2)Mineral dissolution	1)Chemical mechanical weathering	1)Chemical mechanical weathering		1)Ingestion 2)Bioturbation	1)Ingestion 2)Bioturbation	1)Ingestion 2)Utilisation	Interface with Geosphere

Figure 2.2: General matrix representation of the migration of radionuclides in an aquatic ecosystem.

2.3.1. ODEs

Table 2.2 describes the ODEs for aquatic ecosystems. Each transfer process is shown as an ODE in the aquatic matrix. The transfer process definitions are the same as for the terrestrial matrix, see Section 2.1. However it should be noted that the list of processes is not exhaustive in the aquatic matrix.

Table 2.2: Transfer processes included in the ODE of the aquatic interaction matrix.

Physical transport		Chemical interactions	Biological processes	Intake routes
Advection Dispersion Capillary rise Colloid transport Death Degassing Deposition (dry and wet) Diffusion Exhalation	Droplet production Evaporation Excretion Erosion of bed sediment Gas evolution Particle transport Release due degradation Release from solution Respiration Resuspension	Adsorption Carbonate production Chemical weathering Complex formation Desorption Dissolution Fermentation Fixation Isotopic exchange Mineralisation Precipitation Sorption	Biological weathering Bioturbation Excretion Photosynthesis Root uptake Degradation	Inhalation Ingestion Uptake

2.4. Important Processes in Aquatic Environments

Each type of aquatic environment is characterised by specific properties: lakes and lagoons by a low flow regime and variable salinity; rivers by unidirectional and variable freshwater flow regime; estuaries and fjords by bidirectional and variable tidal and freshwater flow regime, and variable and transitional salinity; coastal seas by tidally dominated transport and salinity.

Aquatic ecosystems are also heterogeneous media, composed of: (1) a water column including anything that is dissolved within it (i.e. including colloids < 0.45 µm diameter) and suspended particles (i.e. > 0.45 µm); (2) bed sediment; and (3) benthic and pelagic biota, including primary producers, and primary and secondary consumers.

Generic processes to any aquatic environment are of physical, chemical and biological nature. Physical processes, Table 2.3, include: advection (transport of a solute or suspended particles with the water), dispersion (diffusion), erosion and deposition of particle-bound contaminants (burial/release). Chemical transformations, Table 2.4, are decay and speciation, complexation and degradation (DOC, POC, Fe-Mn oxides), adsorption and desorption. Sorption is influenced by many physical and chemical factors such as salinity, pH, dissolved oxygen, and particle characteristics and concentrations. Biological processes, Table 2.5, are governed by ingestion (of water, sediment and biota) and absorption across body surfaces.

Table 2.3: Physical transport processes in aquatic environments.

Advection and dispersion of solutes	Advection & Dispersion of particulates	Erosion and deposition
High in fluvial, estuarine and fjordic systems (10s of km in a few hours) Moderate in open coastal locations Low in lakes and lagoons	Fine-grained permanently suspended fraction (AD as solute) Temporarily suspended fraction (AD less than solute)	Only relates to temporarily suspended load Current thresholds for deposition/erosion Can be periodic or intermittent

Table 2.4: Chemical transformations in aquatic environments.

Decay and speciation	Complexation and degradation	Adsorption and desorption
Influences sorption (kinetics versus K_ds) Influences bioavailability and toxicity	Influences sorption (kinetics *vs.* K_ds) Influences bioavailability and toxicity	Influences bioavailability and toxicity Influence advection and dispersion Influence (e.g., through burial) speciation/complexation status

Table 2.5: Uptake and exposure pathways in aquatic environments.

Ingestion (particle/solid bound)	Adsorption (dissolved phase)	External exposure from (no assimilation into biota)
Detritus/organic material Primary consumers (filter feeders/ grazers) Secondary consumers Tertiary consumers	Uptake across external body services (gills, mantle cavity, body wall) Uptake across gut wall	Sediment & water external to biota Internal sediment and water (ingested and within the gut, but not assimilated)

Below an overview of the most important processes is provided for each of the studied radionuclides, based on the discussions of the group during the scientific seminars.

2.4.1. ^{14}C

The processes that were considered by the group as most important for describing the ^{14}C behaviour in aquatic ecosystems were: photosynthesis, consumption, respiration, decomposition and excretion by primary producers, herbivorous fish, carnivorous fish and detritivores.

The intake of ^{14}C by fish consumption is often found to be an important exposure pathway. This arises because of the assumption that the specific activity of carbon in fish is equal to the specific activity in the surrounding water, which is often not correct. An improved modelling approach is required that more correctly represents carbon (and hence ^{14}C) fluxes in aquatic ecosystems. This issue is discussed in more detail in Section 3.3.

2.4.2. ^{36}Cl

^{36}Cl is highly conservative in waters and is, therefore, readily transported through the aquatic environment. It may be rapidly dispersed in water bodies or enter the soil system. The movement of ^{36}Cl in water flow through the geosphere is due to advection and longitudinal dispersion along fractures and molecular diffusion into the adjacent rock (Sheppard *et al.*, 1996).

Terrestrial foodchain pathways are thought to be of greater importance than aquatic pathways due to the rapid dilution and dispersion of ^{36}Cl in surface waters and the limited degree of concentration even by freshwater organisms (EA, 2003).

Chloride concentration in aquatic animals and plants is maintained constant by modulating its absorption and excretion rates. As a result, chlorine is ubiquitously present in plant and animal tissues, and is relatively uniformly distributed throughout them.

2.4.3. ^{99}Tc

The main source of Tc in the marine environment is the nuclear fuel reprocessing. The concentrations of Tc coming from reprocessing plants reach several mBq/l, while the contamination with fallout from nuclear tests has resulted in concentrations of 10-20 µBq/l. Technetium exhibits complex chemical, biochemical and biogeochemical interactions. In general, Tc tends to remain dissolved in waters, which makes it an excellent tracer of the movement of water masses.

Technetium behaves generally conservatively in water in the form of TcO_4^-. If Tc remains in oxidised form, it is likely to be highly mobile and reacts only weakly with particulate matter in oxidising conditions (EA, 2003). In anoxic environments technetium is reduced and can be fixed by sediments together with Fe and Mn. Hence, discharges to freshwater, estuarine or marine environments are expected to disperse widely, mixing throughout the world's oceans in the long term. Tc precipitates as hydroxides, oxides and sulphides, and in reducing conditions forms complexes with humic acids. The behaviour of Tc in natural sediments is influenced by complex, often interdependent factors, such as the nature of the microbial population, the types and concentrations of electron donors, the presence of inorganic and organic complexing agents, and the properties of the aqueous and solid phases (Wildung *et al.*, 2004).

Very few living organisms concentrate Tc, although some species of macro-algae, *Fucus sp.* and *Ascophyllum sp.* show concentration factors of 100,000-200,000 (d.w. basis). Aquatic plants differ from terrestrial ones in nutrient uptake, since they can use both foliar and root uptake to a substantial degree. As sediment complexes can be considered as inert and not bioavailable for aquatic macrophytes, foliar absorption of TcO_4^- should be considered as the only route of uptake for Tc in aquatic plants (Hattink, 2000). It is supposed that accumulation of Tc is dominated by two processes:

- transport across the cell membranes (uptake and release of TcO_4^-), and
- photoreduction of Tc in the chloroplast followed by complexation (Hattink,*et al.* 2001).

Tc can be highly available to animals, with fractional gastrointestinal values approaching 100 %. Concentration ratios relative to water for Tc are about 30 for freshwater and marine fish, but are about 1,000 for molluscs and crustaceans. Relatively high concentrations of Tc have been also observed in lobster. A satisfactory explanation for these high concentration factors has not been yet found. Some researches have pointed that these might be a result of biases in the sampling, for example water samples being taken after the passage of the plume. The concentration factors of technetium tend to increase with the salinity, which is in contrast with the concentration factors for caesium, that tend to decrease when salinity increases. This observation could be useful for testing models.

2.4.4. ^{237}Np

In general there are few environmental data for ^{237}Np, due to difficulties of measurement in the past and there are thus gaps in the understanding of its environmental behaviour. ^{237}Np can exist in a number of oxidation states, but only the +4 and +5 states are important in environmental systems. A variety of species in aqueous solution can be formed, depending on Eh and pH.

There are chemical similarities with U and Pu. However, the chemistry, biochemistry and biogeochemistry of ^{237}Np are complex, and neither U nor Pu can be relied upon as quantitative analogues for its behaviour in the environment.

^{237}Np is highly particle reactive in the aquatic environment and therefore tends to migrate to bottom sediments. In aquatic systems, bottom sediments are the most likely environmental sinks and ^{237}Np migration will be closely associated with sediment transport. It is possible that enrichment takes place in specific environments, as is the case for other actinides, especially the redox-sensitive ones such as Pu. Such environments are anoxic water bodies or waters with high humus content. From a waste management point of view such environments are of particular interest.

Concentration ratios of between 10 and 100 are typical for marine and freshwater species.

There is no clear differentiation in the importance of the transfer processes. For some processes, Np mobility seems to be greater than that of Pu and Am.

One area of research that was highlighted was the need to understand the effect of changing redox chemistry on neptunium solubility and mobility in the environment. All the studies thus far point to the fact that neptunium may be more soluble than either Am or Pu in oxidising systems, but that when reduction occurs, Np solubility may decrease. This is particularly important to consider in the context of waste management scenarios.

2.4.5. ^{238}U

There seems to be good knowledge about uranium speciation in solutions. The association between uranium (U-VI) and iron oxide surfaces is also well understood. In general, the interactions in water/sediment and biota/water systems have been quantified systematically in large databases of K_d values and CFs, but a large variability in the values is observed. There are, however, knowledge and data gaps in several aspects of the uranium cycling in aquatic systems, such as the role of sediments as reservoir, the role of colloids, the exchange processes between solution and suspended particles, the role of microbiology and organic matter. It is also necessary to better understand the relationship between speciation, bioavailability and bioaccumulation.

Indeed, chemical processes in the dissolved fraction (<0.45µm) are governed by oxydation-reduction processes, complexation and adsorption to colloids and particles. Existing oxidation states go from +III to +VI and govern the mobility of the element. The dominant oxidation state is U(IV) in confined aquifers and is poorly mobile, whereas U(VI) controls the uranium concentration in oxic environments and is more reactive. The free uranyl ion (UO_2^{2+}) is the dominant species at pH<6, whereas at higher pH, hydroxy- and carbonate-complexes dominate. As direct measurement of speciation at environmental concentration levels is difficult, thermodynamic equilibrium modelling is generally used to estimate the chemical speciation in the presence of various inorganic ligands (OH^-, PO_4^{3-}, CO_3^{2-}) and organic ligands. These calculations require high-quality thermodynamic database, and uncertainties on the reported thermodynamic constants limit the predictive ability of speciation in conditions where several species co-dominate (Denison and Garnier-Laplace, 2005). Other knowledge gaps make uranium's chemical speciation in freshwaters at environmental levels a great challenge:

- complexation of uranium with colloids is poorly characterised and > 90 % of U in water can be linked to iron-rich colloids, humic acids and humins. This fraction decreases with salinity (< 5 % [U]total , S = 3 ‰);
- non-equilibrium systems (e.g. biotic changes and redox state in microenvironments).

Interactions of uranium with sediments and particles (> 0.45 µm) are mainly documented as K_d values. Values are highly variable: the estimated K_d values for uranium go from a maximum at pH = 6 (102 to 106 $L.kg^{-1}$) to a minimum in acidic (< 1 to 32 at pH = 3) and alkaline (< 1 to 5 at pH = 10) sediments.

However, the use of surface complexation models could increase the predictive ability of water/particle exchange of uranium. Important factors influencing the adsorption behaviour of U(VI) on the main active surfaces of particles (organic matter, iron hydroxides, clays) are pH (decreases in pH reduce the number of exchange sites on variable charged surfaces, such as iron-, aluminum-oxides, and natural organic matter) and dissolved carbonates (competition with sorption sites). Precipitation/dissolution also plays a role (precipitation of U(IV) forming uraninite and U(VI) forming schoepite) and in presence of phosphates (formation of ningyoite and autunite solids) and silicates (formation of coffinite and uranophane).

Regarding interactions of uranium with biota, the main identified processes are microbial solubilisation (oxidation of sulphide-ores in aerobic medium by iron/sulphur oxidising and acidophilic bacteria; production of organic acids and ligands: dissolution of silicates, phosphates, carbonates, oxides, sulphides; indirect modifications of physico-chemical conditions) and microbial insolubilisation (direct (bacterial) or indirect (Fe(III)->Fe(II)) reduction of U(VI) to U(IV) in anaerobic conditions (iron-reducing and sulfate-reducing bacteria) ; co-precipitation with iron and sulphur; biosorption / bioaccumulation). Quantification of the relative contributions of the various involved processes and their kinetics is however poor and the controlling factors are not well understood.

Bioaccumulation of uranium in higher organisms is quite well documented, but data show that concentration factors are dependent on the organism's characteristics (species considered, life stage, physiology, etc.), exposure pathways (direct, trophic) and medium characteristics (Quin *et al.*, 1995; Garnier-Laplace *et al.*, 2001). In plants, uranium has a high affinity for proteins and lipids at the intracellular pH. Adsorption at the surface of the cells can be dominant according to the species and primary production can constitute a source of contamination of sediments by deposition and cellular lysis. Few data exist in the literature relating to the contamination of macrophytes (mainly macroalgae). Typical concentration factors ($Bq.kg^{-1}$ ww / $Bq.l^{-1}$ water) are 100 – 1000 for unicellular algae and 4 – 1000 for plants (in general). In animals (crustaceans, molluscs, fish), the bio-concentration factor from water is very low and very few data exist on trophic transfers. Fish concentration factors (direct and trophic) vary from 0.01 to 20 (ww) and values depend on the behaviour of organisms (pelagic species accumulate approximately 10 times less than benthic species) and target tissues (bone [200-8000] and kidneys > liver and gills > muscles [1.5-24.3] > digestive system > gonads).

3. Improvement of the Assessment Models

This section summarises the group's suggestions on ways for improvement of the models of the long-term behaviour of radionuclides in the environment. The sorption to solids of radionuclides and their transfer to biota are among the most important processes affecting the behaviour of radionuclides in the environment. Despite of this, these processes are usually not described explicitly in the models. Instead, distribution coefficients (K_d) and transfer factors (TF), respectively, are used for addressing these processes. The use of the concepts of K_d and TF is often criticised in the literature, but nevertheless they continue to be widely used in assessment models. This is motivated by the simplicity of the approach and because, for both of these parameters, large amounts of data have been collected and reported in the literature. There are, however, disadvantages in using such a simple approach. Obviously, this approach is of limited use when empirical data on K_d and TF values are not available, which is the case for many radionuclides of interest. Also, where data are available, the intervals of variation of the data are often very large, which complicates the selection of appropriate values for a given context. This section discusses ways for dealing with these disadvantages and alternative approaches for the parameterisation of the underlying processes. The section also includes a discussion on dynamic models of the environmental behaviour for ^{14}C, which is one of the radionuclides for which the concepts of K_d and TF are least applicable.

The section concludes with a discussion on two more issues that were briefly addressed by the group: 1) potential problems with the use of compartment models for describing the transport of radionuclides, 2) modelling the input of radionuclides to the biosphere with groundwater.

3.1. Improvement of the description of sorption processes

3.1.1. Introduction

Soil K_d values, or solid:liquid distribution coefficients in units of L/kg, sometimes called partition coefficients, continue to be used in safety assessments worldwide. They are used in hazardous substance assessments as well as in nuclear-operations and nuclear-waste-disposal assessments and impact statements. Many papers have been written discouraging their use, but despite that, default values are widely used (Sheppard and Thibault, 1990) and useful in scoping assessments and they are relatively easy to measure if site-specific data are required. Specific processes occurring at the liquid-solid interface are difficult to pinpoint and if knowledge of what these processes are is not important, then a K_d value can be useful and appropriate.

There are several types of uncertainties associated with Kd data: they include lack of knowledge about the element speciation in the solution contacted with the solid/porous media, insufficient ancillary data describing the solid media and sometimes poorly described or incorrect protocols used to determine the K_d value. In addition, if the values are a result of a laboratory experiment, there are the uncertainties or difficulties of relating a value produced in the laboratory with those conditions relevant to the field or application context. Also, there are the uncertainties of applying the data from a small subsample of a whole system to the whole system, i.e. redox conditions will differ, microbial populations will vary, moisture will be lost, etc. Other uncertainties with the use of K_d values arise when the K_d values do not represent equilibrium conditions, or difficult-to-control speciation changes occur during the measurement process.

Several other measurement-related errors or uncertainties can also arise and are discussed. The relationship between the soil K_d value and the soil ancillary properties is briefly discussed. Uncertainties related to analytical protocols and measurements of element or radionuclide concentrations or activities are not discussed here.

3.1.2. Subsampling

As with all environmental sampling, there is always an issue with how well the subsample, if small – a few tens of grams - represents the whole soil or sediment. Multiple samples are always better to represent the entire system and cover the spatial variability found in the field. However, describing this spatial variability among the samples, how they differ physically, chemically and where they were sampled with respect to one another and reporting a Geometric Standard Deviation (GSD) although helpful is not always done. If a soil or sediment is very heterogeneous, then reporting the individual data, along with their physical and chemical attributes and a combined site value might be more appropriate. Several papers have shown substantial changes in soil K_d values along a transect or spatial tangent (Bunzl and Schimmack 1989, Sheppard *et al.*, 1989b).

3.1.3. Ancillary Data

The importance of the knowledge of ancillary media properties is emphasised. These include bulk density, texture including individual grain size fraction (%sand, %clay), organic matter or organic carbon content, soil or sediment pH, sampled moisture content, soil field capacity moisture, concentration and composition of the contacting solution, mineralogy of the solids and some knowledge of the expected speciation of the metal or radionuclide of concern. For some elements, such as Cs, Cl, I and C, it is important to know the background or stable element concentration, since this either affects the sorption process or is required for a Specific Activity type model.

A thorough description of all the experimental details and details of ancillary data is very important for those trying to correlate the soil properties with the K_d value or apply the data to similar soils or sediments. Soil pH, clay and organic matter contents as well as mineralogy have all been shown to be important for determining soil K_d for a variety of metals and radionuclides. Also, soil pH and organic matter content can vary significantly with time depending on soil management and fertilisation practices.

For uranium, one of the most recent studies to investigate the effects of soil characteristics on the K_d value is Echevarria *et al.* (2001). They found no significant effect of clay or organic matter content; however, they did find a significant relationship between soil K_d (as L kg^{-1}) and pH. For 13 soils, the relationship was

$$\log K_d = a\,pH + b \qquad (3.1)$$

where $a = -1.25 \pm 0.13$ (standard error) and

$b = 10.9 \pm 1.0$ (standard error).

The r^2 value was 0.894. They obtained Kd values for eight additional soils using the same experimental conditions as in the Canadian work (Sheppard *et al.*, 1989a) and found the relationship was not changed significantly with $a = -1.29 \pm 0.17$ (standard error) and $b = 11.0 \pm 1.2$ (standard error), n=21, and the r^2 value was still significant at 0.755. This significant relationship with soil pH is probably due to the existence of different uranium complexes as a function of soil pH. Echevarria *et al.* (2001) stated that soil pH should be the focus variable for reduction of uncertainty associated with soil K_d for risk assessments.

The soil pH range of Echevarria *et al.* (2001) was from 5.5 to 8.8 with a data set of 21 values. The compilation of soil K_d values used here includes 134 data. Some of these data did not have accompanying pH data. Further, because Echevarria *et al.* (2001) did not include organic soils, our data set was reduced to 119 values and produced the following regression equation:

$$\log Kd = -0.486\ pH + 2.56 \qquad (3.2)$$

The parameters of this equation are significantly different from those of Echevarria *et al.* (2001), and the overall r^2 for this compilation ($r^2 = 0.18$) is much poorer. This can be partially explained by the larger range of soil pH in this compilation – from 4.1 to 8.8 as opposed to 5.8 to 8.8 for Echevarria *et al.* (2001). Also, this compilation contains sorption values from many different measurement protocols, whereas Echevarria *et al.* used one consistent protocol. Figure 3.1 shows the plot of soil Kd versus pH for this data compilation and its relationship for all soils including the regression line of Echevarria *et al.* (2001).

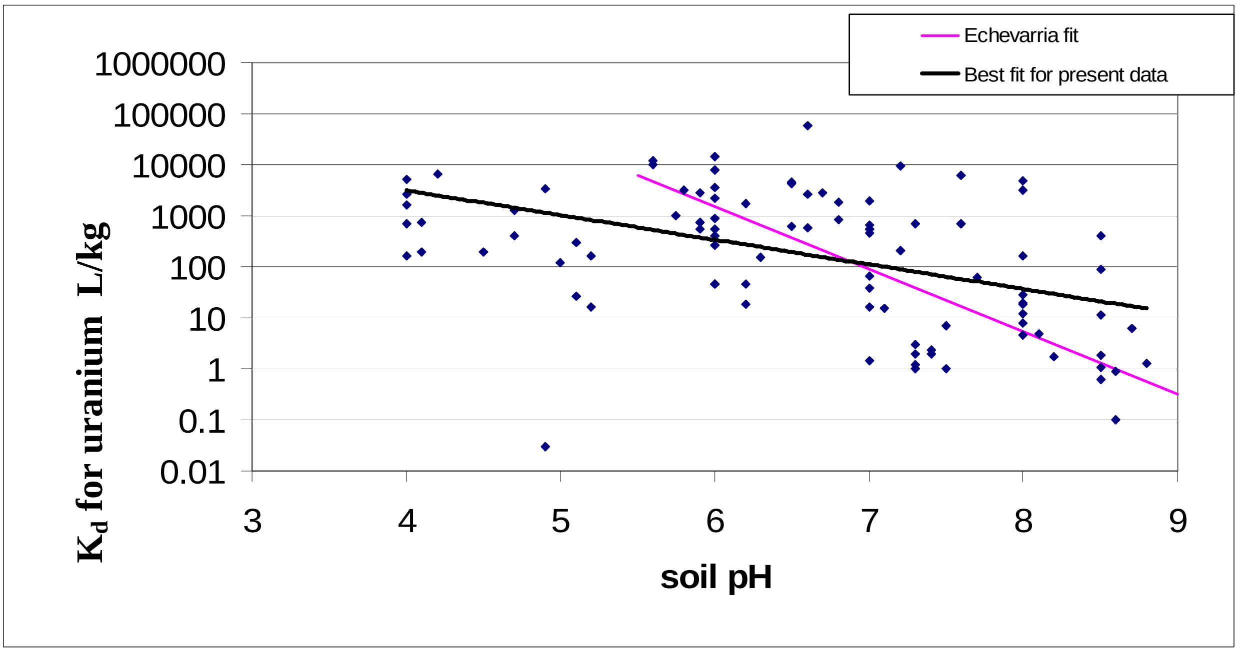

Figure 3.1: Soil Kd versus pH for this data compilation and its relationship for all soils including the regression line of Echevarria *et al.* (2001).

Ancillary data are also important when linkage to other parameters such as bio-uptake and toxicity are required. In these cases, correlations to other attributes of the system should not be lost.

3.1.4. Sample Preparation

Sample preparation uncertainties start with how the sampler treats the soil or sediment from the time it is removed from the site through how it is preserved during its journey back to the laboratory (sealed or open, upright for sediment cores, etc.) to how it is treated once it is in the laboratory (refrigerated, frozen) and how long it is stored before any experimental work begins. More uncertainties are associated with drying and grinding (are bigger stones or rocks removed?) and some protocols use a particular soil or sediment size fraction rather than the whole soil or sediment. Also, a flush of microbial activity can occur on rewetting a dried soil. It is important to consider how all of these factors could influence the final K_d value. The application context is important, for example is the K_d value required to be representative of agricultural soil at field capacity, under fully aerated conditions or fully saturated. Batch sorption protocols (Roy *et al.* 1991) usually work at a 1:10 soil to solution ratio and this is more appropriate for sediments and subsoils than for surface agricultural or urban soils.

3.1.5. Measurement Protocol

If the method used to produce a K_d value is for sorption as opposed to desorption and analysis of the moisture from the soil or sediment sample as it was in-situ (appropriate for contaminated sites), then it is important to understand the effects of the soil:solution ratio on the element or radionuclide of interest. This ratio affects the soil solution composition (Skyllberg, 1995) and metal desorption (Yin *et al.* 2002) and hence the resulting K_d value. Redox, pH and element speciation changes during the experiment could cause spurious results leading to uncertainties in the data. The soil gas composition may change and, if the partial pressure of CO_2 is important, as in the sorption of ^{14}C, this leads to uncertainties or bias in the data. Sorption experiment times range from a few hours to months. The underlying assumption of the use of the K_d value is that the value is obtained at equilibrium, and this needs to be demonstrated.

Removal of liquid, often carried out at intervals to get a time sequence for demonstrating the equilibrium condition, needs to be small enough to not upset the soil:solution composition or remove substantial amounts of the sorbing element. Filtering of the liquid is always a concern; does it remove any colloidal or very fine particulate that should really be considered part of the solid media? Analysis of the solid, if required to get the concentration on the solid phase, would probably include a chemical digestion and uncertainties arise as to what fraction of the total solid any digestion accesses, or what particular solid phase any chemical extraction represents. Sorption by difference is problematic for soluble elements. For example, if the detection level in the solution is only accurate to ± 1 % and less than 1% is sorbed, detection errors become problematic. Also, if the solid concentration is required, one must account for the element associated with the liquid that is dried onto the solid and left behind with that phase.

3.1.6. Data Analysis

Poor detection or very low concentrations in the liquid phase could lead to negative K_d value estimates, perhaps more problematic when the solid concentration is determined by subtraction. This has been an issue for Tc (Sheppard *et al.*, 1989b).

Another concern is how to quantify uncertainty in final numbers. Since the K_d value is usually lognormally distributed, the Geometric Mean and Geometric Standard Deviation are the appropriate terms to use. An example of how the soil K_d value for U is distributed is shown below in Figure 3.2.

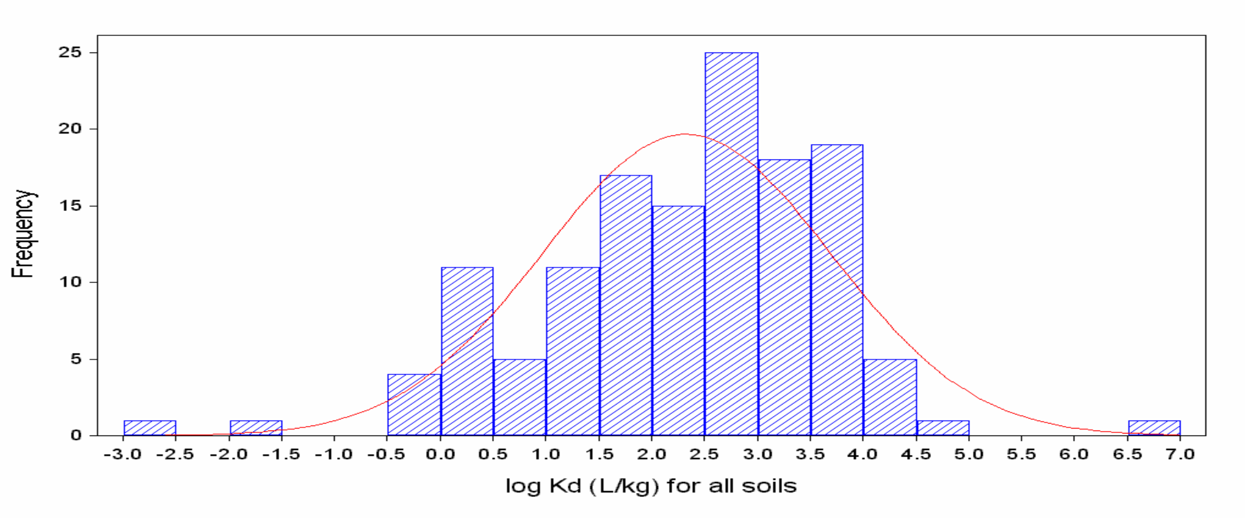

Figure 3.2: Histogram of all soil K_d values for uranium. The smoothed curve is the best-fit lognormal distribution (Sheppard *et al.* 2005).

3.2. Improvement of the description of transfer to biota

To estimate radionuclide concentrations in biota, the most commonly used method is to multiply the concentration in soil or water by a transfer factor (TF). This method is easy to use and there exist compilations of TFs for many radionuclides. However, the TF values have a large variability, as they are influenced by several chemical, biological, hydrological, physical and physiological processes. Also, the TF values are of limited use when there are no or few data available, as they cannot be easily scaled from one situation to another. Here we provide some suggestions on methods for filling data gaps and reducing uncertainties in TF values from soil-to-plant and to fauna. The specific problems associated with the application of TF values for calculations of the concentrations of ^{14}C in biota, and possible alternative solutions to these problems, are discussed in Section 3.3.

3.2.1. Parameterisation of soil to plant transfer factors using mechanistic models

The soil-to-plant transfer factors, often called Concentration Ratios (CRs), relate the concentration in the plant (expressed in Bq/kg DW), to the total concentration in the soil, or in given layer of the soil (expressed in Bq/kg DW). This definition implies that there is a linear relationship with zero intercept between the total radionuclide concentration in soil and plants, which is not always the case (Sheppard and Evenden, 1988). Moreover, the radionuclide uptake rates are often proportional to the available fraction, rather than to the total radionuclide concentration in the soil (Desmet *et al.*, 1991). As a result, a large variation in the concentration factors is commonly observed. The above shortcomings can be partly overcome by using probability distributions, instead of single values, for the concentration ratios. It has been also proposed to assume a negative correlation between the concentration ratios and the radionuclide distribution coefficients in soil (Sheppard and Evenden, 1988; Sheppard and Sheppard, 1989), although the correlation coefficients show substantial variability (Alexakhin and Krouglov, 2001). Another problem with the use of CR values is that for many radionuclides of interest values are lacking. Some of the data gaps could be filled by performing site-specific measurements of the radionuclide concentrations, or the corresponding stable analogues in soil and vegetation. However, it should be taken into account that CR values obtained for the stable analogue and the radionuclide may differ, for example if their availability for uptake by plants is different.

In the frame of the EC project BORIS (Tamponnet *et al.*, 2001) an approach for deriving expressions relating the CR values to soil and plant parameters was developed (Norden *et al.*, 2004). This approach was applied for derivation of CR values used in a forest model (Avila, 2006) with satisfactory results. The essence of the approach is to express explicitly the radionuclide transfer to plants as a function of the bioavailability of the radionuclide in soil. The term bioavailability has been widely used in radioecology, not only for description of soil-plant interactions, but also to describe transfer to animals. However, there is a lack of a widely accepted definition of bioavailability that is both theoretically grounded and operational.

The following definition of bioavailability is proposed in (Norden *et al.*, 2004): *the bioavailability of a radionuclide in soil is a measure of the potential for the radionuclide in soil to be taken up by plants within a given time interval (for example within one vegetative period for perennial plants or within one growing season for crops)*. Further, to make this definition quantifiable, the potential for being taken up by plants can be interpreted as the probability that the radionuclides are taken up by plants within a given time interval, which can only take values between 0 and 1. The total bioavailability corresponds to the total, unconditional, probability, which equals the sum of the values of the bioavailability of the radionuclide in different soil pools multiplied by the fraction of the radionuclide in the corresponding pool. From this definition, a simple equation for the CR as a function of the bioavailability can be obtained. The bioavailability can then be estimated from experimental studies or with the help of mechanistic models as shown in (Norden *et al*,. 2004). For the case when the total bioavailability is approximately equal to the fraction of radionuclides in the soil solution and if there is no substantial radionuclide depletion near the roots, then a simple equation for the bioavailability is obtained, which yields the following equation for the CR:

$$CR = \frac{Depth * \theta}{Biomass * (1 + F) * Kd} \tag{3.3}$$

where,

Density - is the bulk density of the soil rooting layer [kg/m^3]

Depth - is the depth of the soil rooting layer [m]

Biomass - is the plant biomass [kg/m^2]

K_d is the distribution coefficient for the rooting layer of the soil [m^3/kg]

θ is the volumetric water content in the rooting layer of the soil [m^3/m^3]

F is a correction factor, equal to the ratio of the radionuclide activity in the below-ground and above-ground parts of the plant.

The above equation indicates that for the assumed conditions the CRs are inversely proportional to the distribution coefficients. This seems to be consistent with observations found in the literature for some radionuclides under certain conditions (Sheppard and Evenden, 1988; Sheppard and Sheppard, 1989). Figure 3.3 shows results of preliminary tests of the validity of this relationship for prediction of CR values for forest understorey plants using data presented in Avila (2006). This simple expression can capture the overall trend and performs well for some elements, for example Cl, Sr, Ni, U and the actinides. However, there is substantial disagreement for some elements, like Th, Ra, Cs, I and Tc. It should be taken into account, that the literature data used in the comparison were obtained at different sites and using different methods. A better agreement could be achieved if the model parameters and the empirical data on CR values were obtained for the same site. Hence, it can be concluded that this approach has good prospects of being useful, in particular for radionuclides that are not analogues of plant nutrients and also some analogues of plant micronutrients.

As shown in (Norden *et al.*, 2004), it is relatively easy to parameterise the bioavailability, when defined as above, even when using more sophisticated mechanistic models that take into account relevant processes for each specific radionuclide, including interactions between the radionuclide and the stable analogues. This would lead to more of less complex expressions relating the CR values of the different elements with key soil and plant parameters. In situations where the use of CR values is not appropriate, it is possible to use the parameterisation of the bioavailability to obtain parameterisation of the radionuclide fluxes that can be used directly in dynamic models (Norden *et al.*, 2004 and Avila, 2006).

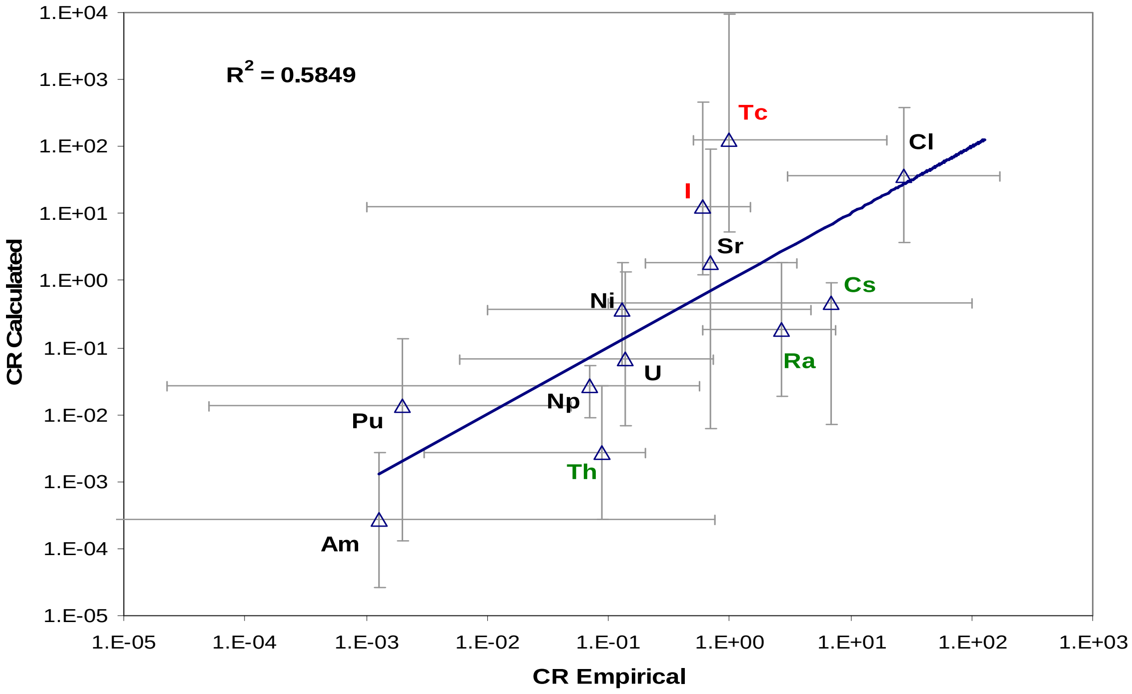

Figure 3.3: Comparison of empirical values of Concentration Ratios (Bq/kg DW per Bq/kg DW) from soil to forest understorey plants against values calculated with equation (3.3).

3.2.2. Use of kinetic-allometric models for deriving transfer factors to fauna

For several radionuclides of interest in the context of the management of HLW, there is lack of empirical data on TF values to fauna, in particularly for wild animals. A possible approach for filling these gaps is to derive the TF values from kinetic models where some of the parameters are expressed as functions of the weight or size of the animals - allometric equations (Highley *et al.*, 2003; Avila, 2006). Depending on the radionuclide, more or less sophisticated kinetic models of metabolism in animals can be applied. For example, using this approach, an allometric equation was obtained in Avila (2006) for a transfer factor (TF) that relates the activity concentration in the whole body of mammals to the activity concentration in the animal diet (expressed in Bq/kg FW per Bq/kg DW):

$$TF = f \quad * a * W^{b^{j}} \qquad (3.4)$$

where,

f is the gut uptake fraction of the radionuclide [-]

a is the multiplier in the allometric relationship for the ***j-th*** radionuclide [in appropriate units]

b is the exponent in the allometric relationship for the ***j-th*** radionuclide [-]

W is the weight of the animal [kg]

The values of the parameters *a* and *b* in Table 3.1 were derived from the kinetic allometric model as described in Avila (2006). Several allometric relations were used in the derivation including: the biological half life of the nuclide (Beresford *et al.*, 2003, FASSET, 2003, Highley *et al.*, 2003), the life span of the animal (Calder, 1984) and the dry matter intake (Nagy, 2001).

Table 3.1: Values of the parameters used in the allometric equation of the TF (equation 3.4); *a* is the multiplier in the allometric equation (in appropriate units), *b* is the exponent in the allometric equation (r.u.) and *f* is the gut uptake fraction (r.u.).

Nuclide	a	b	f	References
Am	2.0E+1	1.4E-1	5.0E-4	Coughtrey *et al.* (1985), Beresford *et al.* (2000)
Cl	2.1E-1	1.1E-2	1.0E+0	Bishop *et al.* (1989)
Cs	1.2E+0	0.0E+0	1.0E+0	Coughtrey *et al.* (1985), Beresford *et al.* (2000)
I	1.4E+0	-1.1E-1	1.0E+0	Beresford *et al.* (2000)
Ni	2.2E+1	1.1E-1	5.0E-2	Coughtrey *et al.* (1985)
Np	2.2E+1	1.1E-1	1.0E-3	Coughtrey *et al.* (1985)
Pu	2.0E+1	1.4E-1	5.0E-4	Coughtrey *et al.* (1985), Beresford *et al.* (2000)
Ra	1.4E+1	8.0E-2	2.0E-1	FASSET (2003)
Sr	1.8E+1	8.2E-2	2.0E-1	Coughtrey *et al.* (1985), Beresford *et al.* (2000)
Tc	4.2E-1	1.6E-1	1.0E-1	Bishop *et al.* (1989)
Th	1.8E+1	1.3E-1	2.0E-4	FASSET (2003)
U	4.8E-1	4.1E-2	5.0E-2	FASSET (2003)

The values of the allometric coefficients ***a*** and ***b*** were derived in Avila (2006) using a kinetic-allometric model. The references refer to the gut uptake fraction (*f*).

3.3. Modelling the long-term behaviour of ^{14}C in terrestrial and aquatic environments

3.3.1. Introduction

As noted in Section 2, post-closure radiological assessments of solid radioactive waste disposal have shown that the generation of ^{14}C-methane and ^{14}C-carbon dioxide can be of radiological significance. This is particularly the case for low and intermediate level radioactive wastes, so the evolution, transport and radiological impact of bulk and trace gases containing ^{14}C has been extensively studied in this context, notably by United Kingdom Nirex Limited (Nirex).

An overview of the behaviour of ^{14}C in gaseous form evolved from low and intermediate level wastes being considered for disposal by Nirex has been provided by Thorne (2005a). In the period immediately after emplacement of ^{14}C containing wastes, the repository will be ventilated and ^{14}C released from the waste packages will be discharged with the ventilation air. The forms discharged are likely to be both $^{14}CO_2$ and $^{14}CH_4$, as even though oxic conditions would prevail in the repository as a whole, anoxic conditions could develop within some waste packages. As the discharges would be to the above-ground atmosphere, standard techniques for atmospheric dispersion modelling can be applied. However, issues then arise relating to the interactions of the dispersing plume with the ground and transfers of ^{14}C from the plume to plants and animals. Interactions of a dispersing methane plume with soils and plants are discussed in Section 3.3.2, whereas interactions of ^{14}C-labelled carbon dioxide with soils and plants, whether entering by discharge from the geosphere or due to dispersion of an atmospheric plume, are discussed in Section 3.3.3.

In the post-closure phase, peak bulk production rates for hydrogen in the proposed Nirex repository could be some tens of thousands of cubic metres per year and peak bulk production rates of methane could be a few thousand cubic metres per year. The magnitude of these production rates is such that the bulk gas is likely to move away from the repository in the free gas phase. Explicit calculations suggested that the timescale for the gas pathway to form could be a few thousand years, but this estimate is not very robust.

The main ^{14}C-labelled gases potentially available for transport away from the repository are likely to be ^{14}C-labelled methane from degradation of organic materials and from metal corrosion, and ^{14}C-labelled acetylene, ethylene and ethane from metal corrosion (notably from Magnox). Other hydrocarbons are likely only to be produced in very small quantities and carbon dioxide is likely either to be involved in carbonation reactions removing the ^{14}C to the solid phase or to react with bulk evolved hydrogen to produce methane.

Although, in principle, some microbial metabolism of methane, acetylene, ethylene and ethane could occur in the geosphere, this cannot be relied upon. However, the potential microbial metabolism of these compounds on entering the soil zone is of considerable significance in evaluating the radiological impact of their ^{14}C-labelled forms.

In addition to being released in gaseous form, ^{14}C may be released from a repository in solution in groundwater.

Thus, in summary, the various forms of ^{14}C likely to be released from a radioactive waste repository include:

- ^{14}C-labelled carbon dioxide, methane, acetylene, ethylene and ethane;
- ^{14}C dissolved in groundwater.

Having been transported through the geosphere, ^{14}C in gaseous form or dissolved in groundwater may be released to soils, sediments or surface water bodies (both freshwater and marine).

In this section, no attempt is made to discuss all issues relating to the modelling of ^{14}C transport in the environment. Rather, specific attention is given to recent developments that are specifically relevant to modelling the radiological impacts of releases of ^{14}C from repositories for solid radioactive wastes.

3.3.2. Modelling of atmospheric dispersion of ^{14}C-methane

By explicit calculations under typical UK atmospheric stability conditions, Thorne (2006b) has been shown that deposition velocities of less than 1 10^{-4} m s^{-1} give very little reduction in the concentration of methane in a dispersing atmospheric plume out to 10 km downwind. Although demonstrated in a specific plume model, similar results would be expected for other, more complex dispersion models.

Thorne and MacKenzie (2005) summarised a variety of data on the rate at which methane is metabolised in the soil zone. These data are reproduced in Table 3.2.

Table 3.2: Rates of methane metabolism in soils.

Consumption ($mg\ m^{-2}\ d^{-1}$)	**Setting**	**Location**	**Reference**
0.2 to 4.9	Temperate forest	New York	Crill (1991)
0.04 to 1.1	Mixed forest	Ottawa	Lessard *et al.* (1994)
0.3	Agricultural soil	Ottawa	Dunfield *et al.* (1995)
0.48 to 0.84	Grass pasture	Colorado	Mossier *et al.* (1991)
0.29 to 0	Agricultural soil	Ottawa	Lessard *et al.* (1997)
0.5 to 0	Agricultural soil		Hansen *et al.* (1993)
1.2 to 4.8	Mixed forest	Pennsylvania	Bowden *et al.* (2000)
1.8	Forest soil	Norway	Jensen and Olsen (1998)
0.014	Arable soil	Norway	Jensen and Olsen (1998)
0.25 to 3.6	Temperate forest	Germany	Born *et al.* (1990)
3.5 to 4.2	Forest soils	New England	Steudler *et al.* (1989)
0.27 to 1.57	Coniferous forest	Northern Quebec	Adamsen and King (1993)

The data in Table 3.2 only show oxidation (consumption) of methane. Soils high in organic matter and poorly drained are a net source of methane, and so would have a negative oxidation (consumption) rate.

There is a very distinct difference in oxidation rate between forest and disturbed soils. Disturbed soils, such as agricultural soils have a much lower oxidation rate. Thus, one study reported a 130-fold lower rate of oxidation in an arable soil compared with an adjacent forest soil.

Sheppard (S C Sheppard personal communication to M C Thorne, August 2004) estimated that the geometric mean consumption rate of methane in soils relevant to Ontario power reactor sites is 0.3 mg m^{-2} d^{-1}. Excluding anaerobic soils, in which no metabolism of methane to carbon dioxide would be expected, a range of consumption rates from around 0.014 to 4.9 mg m^{-2} d^{-1} has been observed, with some indication that the lowest values occur in agricultural soils.

These data on consumption rates can be used in conjunction with the concentration of methane in the atmosphere to estimate a deposition velocity for methane. For this, it is assumed that the concentration of methane in the above-ground atmosphere is identical to the concentration in the soil atmosphere. This is justified by the rapid exchanges that take place between these components of the system.

The globally averaged atmospheric surface abundance of methane in 1998 was 1.745 ppm (IPCC, 2001). Spatial and temporal distributions of tropospheric and stratospheric methane have been determined by extensive high-precision measurements and the range of tropospheric variation is small, but there is a long-term increasing trend, with the atmospheric concentration having risen from around 1.63 ppm in 1985 to 1.72 ppm in 1990 and 1.75 ppm in 2000 (IPCC, 2001). Here, a rounded reference value of 1.8 ppm is adopted.

The air concentration is given on a volumetric basis and corresponds to a concentration of 8.04 10^{-5} mole m^{-3} or 1.29 10^{-3} g m^{-3}. Thus, for a metabolic rate of 0.3 mg m^{-2} d^{-1}, the implied deposition velocity is 3 10^{-4}/1.29 10^{-3} = 0.2356 m d^{-1}. This corresponds to 2.7 10^{-6}

m s^{-1}. For the range of metabolic rates of 0.014 to 4.9 mg m^{-2} d^{-1}, the corresponding range of deposition velocities is 1.3 10^{-7} to 4.4 10^{-5} m s^{-1}.

It is instructive to compare these implied deposition velocities with the deposition velocity for the more reactive gas methyl iodide. This is of considerable interest in the context of releases of ^{131}I from nuclear installations and has been extensively studied. Observed deposition velocities are typically around 1 10^{-4} m s^{-1}, with an estimated uncertainty factor of 5 to 10 in either direction (Thiessen *et al.*, 1999), i.e. 1.0 10^{-5} to 1.0 10^{-3} m s^{-1}. The range of deposition velocities for methyl iodide is higher than that estimated for methane (above), as would be expected given its higher reactivity.

From this analysis, Thorne (2005b) concluded that plume depletion due to methane utilisation in the soil can be neglected in assessing the radiological impact of releases of ^{14}C-methane.

Because deposition is due to oxidation of ^{14}C-methane in the soil zone, this corresponds to direct entry of ^{14}C-carbon dioxide into the soil zone. Modelling of this route of exposure is discussed in Section 3.3.3.

3.3.3. Modelling the behaviour of ^{14}C-labelled carbon dioxide in soils and plants

Various models for the transport of ^{14}C in terrestrial and aquatic ecosystems were reviewed in BIOPROTA (2005). However, few of those models addressed details of the transport of ^{14}C in soils following its entry in groundwater or in a flow of gas originating from a repository. However, Nirex has developed a detailed model of ^{14}C entering the soil zone in gaseous form and the UK Food Standards Agency (FSA) has developed a similar model for representing the behaviour of ^{14}C in solids from sewage treatment used for land amendment. These models have been compared in Thorne (2005c).

Both Nirex and the FSA use multi-compartment models with similar overall structures. However, for chronic releases of ^{14}C to the soil zone, the Nirex model was found to give much lower specific activities in crops than the FSA model (Thorne and MacKenzie, 2005). Inspection of the FSA model showed that this arose from the assumed effectiveness of plant accumulation of ^{14}C from the sub-canopy atmosphere, i.e. the region between the ground surface and the average height of the plant cover. This further followed from the assumption that most plant carbon is derived from the sub-canopy atmosphere rather than the above-canopy atmosphere.

The Nirex results derived from studies with an empirical parameterisation of results from the detailed compartmental model RIMERS. The general structure of the RIMERS model had been documented in public domain literature (Nirex, 1994). Like the FSA model (Thorne *et al.*, 2003), it includes competition between plant uptake from the below-canopy atmosphere and transfers to the above-canopy atmosphere. However, the parameter values adopted for these transfers had not been documented in any public domain literature. It seemed likely that the substantial difference between the results reported related to substantially different assumptions about the effectiveness of carbon dioxide uptake from the below-canopy atmosphere relative to losses from the below-canopy to the above-canopy atmosphere. However, in the absence of detailed data on the RIMERS model and its parameterisation, it was not possible to confirm this speculation.

In order to investigate this issue further, Nirex staff recovered the relevant papers from their archives and made them available to Mike Thorne and Associates Limited. This allowed a detailed comparison of the RIMERS model with the more recent FSA model.

The overall structure of the RIMERS model, which relates only to soils and plants, is shown in Figure 3.4. The model is compartmental in nature and assumes a time-invariant stable carbon content in each of the compartments. The FSA model is also compartmental in nature, but includes soil, plants and grazing animals, see Figure 3.5.

Details of the parameterisation of the two models are given in Thorne (2005c). From a detailed comparison of the two models, it was found that the key disagreement between the models is whether exchanges between the soil solution, soil atmosphere, below-canopy atmosphere and above-canopy atmosphere are sufficiently rapid for equilibrium to be achieved in the presence of a rapid advective removal process from the above-canopy atmosphere. This matter could not be explored in the original RIMERS framework, so an enhanced version of the model was implemented in which these exchanges were represented explicitly. This re-implementation did not affect the compartmental structure shown in Figure 3.4. Rather, it was required because some of the compartments were originally treated as being in equilibrium with each other and exhibiting the same specific activity.

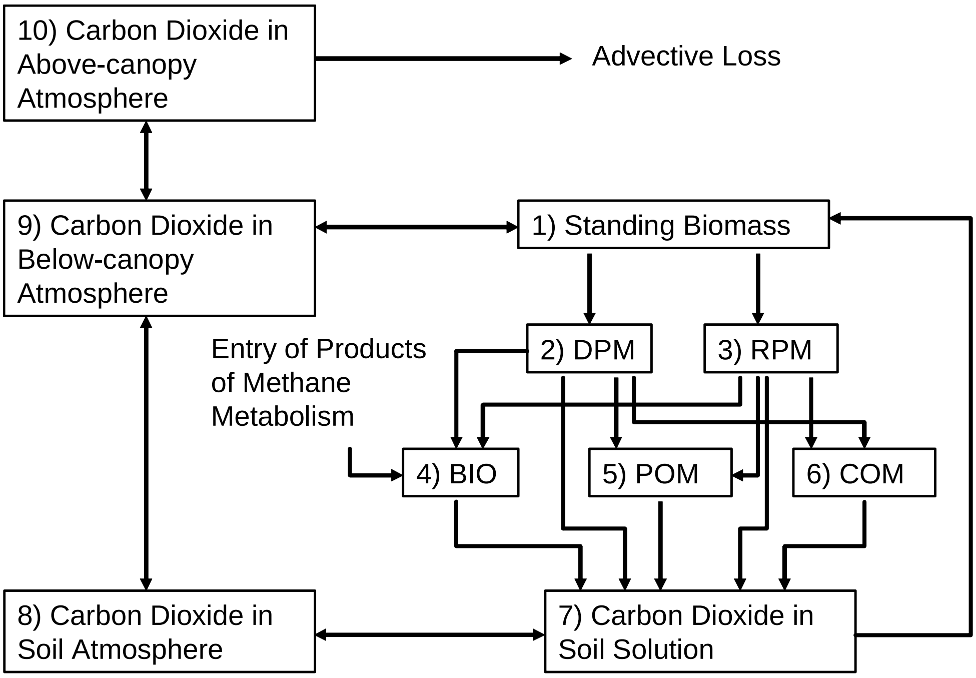

Figure 3.4: Structure of the RIMERS Model.

Overall, it was found that the enhanced RIMERS model gave results in good agreement with the FSA model when the key parameters of the models are tuned to match each other. Having demonstrated this consistency, detailed sensitivity studies were undertaken with the enhanced RIMERS model (Thorne, 2005c and, more particularly, Thorne, 2006).

In the detailed sensitivity study (Thorne, 2006), it was recognised that, because of the need to maintain detailed mass balances for all the components of the model, the input parameters could not be treated as independent. Therefore, a conventional single parameter sensitivity study, in which each parameter in turn is varied about its reference value with the other parameters held fixed at their reference values, could not be undertaken. Instead, each process represented in the model was considered in turn and joint variations in the parameters affecting that process were studied, with each parameter value combination specifically selected to maintain mass balances in the model. Finally, on the basis of these process-based sensitivity analyses, a set of sensitivity calculations was undertaken to explore the effects of sensitivities to various processes in combination.

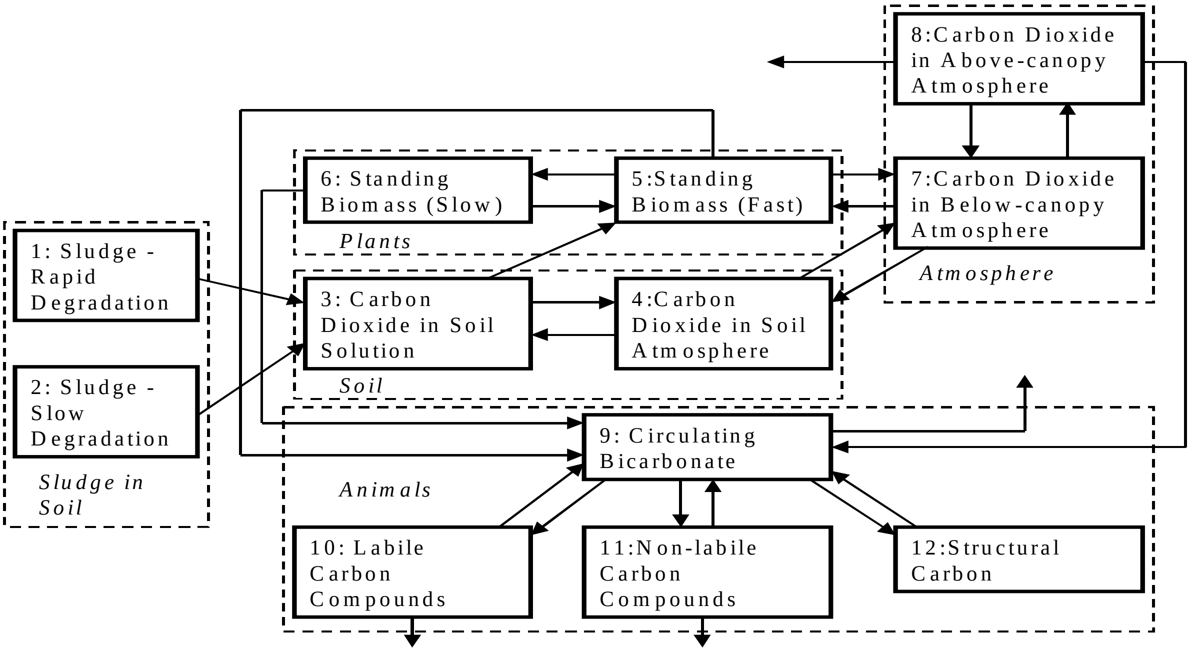

Figure 3.5: Structure of the UK Food Standards Agency (FSA) model.

Overall, it was demonstrated that:

- results of the model were robust against alternative assumptions as to release area and meteorological conditions;
- making self consistent changes of throughput of biomass and uptake of carbon dioxide from the below canopy atmosphere made little difference to the concentration of ^{14}C in biomass in the long term, though the kinetics of approach to the long term equilibrium value were substantially altered;
- model results were robust against changing rate coefficients for equilibration between soil solution and soil atmosphere;
- model results were somewhat sensitive to sensitive to exchange rates between the soil atmosphere and the below canopy atmosphere, but, overall, the effects of uncertainties in the exchange rate between soil atmosphere and below canopy atmosphere were thought likely to be limited;
- biomass concentrations of ^{14}C could be considerably increased if exchange rates between the below canopy atmosphere and above canopy atmosphere were substantially reduced below those assumed in the reference case, but it seemed more likely that, for pasture and arable crops, exchange rates more rapid than those used in the reference case would be encountered and that these could reduce concentrations of ^{14}C in biomass by as much as a factor of 5 relative to the reference case;
- changes in the balance between photosynthesis and respiration in plants (assimilation efficiency) were found to have only very limited effects over a realistic range of assimilation efficiencies;

Sensitivities to combined processes could largely be explained as being due to the multiplicative effect of the individual factors. In some cases, the factors combined significantly less than multiplicatively and in a few instances the effect was slightly greater than multiplicative, but by no more than about a factor of four.

Overall, the various studies undertaken indicated that ^{14}C concentrations in vegetation for chronic exposure are unlikely to be more than a factor of five less than in the reference calculation, which was based on parameter values that were regarded as realistic, and could be up to about a factor of ten higher. Concentrations up to a factor of about 100 higher than those obtained in the reference calculation were obtained in some extreme cases, but these were not considered likely to be realised in practice.

The FSA model and also the enhanced RIMERS model were formulated primarily to address releases of ^{14}C to the soil zone. However, they are also suitably structured for calculating the radiological implications of releases to atmosphere. Illustrative calculations using the enhanced RIMERS model for a spike release to the above-canopy atmosphere are provided in Thorne (2005c).

Also, the models are suitable for use in relation to discharges to the soil zone as gaseous carbon dioxide or methane, or dissolved forms of these gases, as rapid equilibration between soil solution and the soil atmosphere can be expected to occur for either form of release.

3.3.4. Behaviour of methane, acetylene, ethylene and ethane in the soil zone

Some comments are provided on the behaviour of methane in the soil zone in Section 3.3.2, where rates of oxidation are discussed. Thus, in some soil contexts, ^{14}C-methane will be oxidised to ^{14}C-carbon dioxide, which can be modelled as described in Section 3.3.3. Detailed information on the likely behaviour of acetylene, ethylene and ethane in the soil zone is given by Thorne (2005d). In summary, although acetylene may be metabolised to ethylene in some soil contexts, there seems to be no reason to suppose that any significant fraction of ^{14}C entering the soil system as acetylene, ethylene or ethane would be either retained in the soil or taken up by plants. Therefore, the view of Rees *et al.* (2003) is confirmed that radiological impacts of releases of these gases will be determined by the inhalation pathway.

3.3.5. Uptake and retention by animals

Some ^{14}C released as radioactive gas will be incorporated in plant materials and taken up by animals. A wide variety of studies on the biokinetics of uptake, retention and excretion of ^{14}C in and by animals has been undertaken. An integrated model for uptake by animals in an agricultural context has recently been described by Maul *et al.* (2006). Recently, a blind validation study of this animal model has been undertaken (Thorne *et al.*, 2006).

The model structure adopted for both ^{3}H and ^{14}C is illustrated in Figure 3.6. Ingested ^{14}C is partitioned between a highly labile bicarbonate pool, from which rapid losses occur mainly by exhalation, and two organic pools that exhibit turnover at different rates. Timescales of carbon turnover range from minutes in the bicarbonate pool to hundreds of days is the non-labile organic pool. Indeed, this model is a considerable simplification, and many carbon pools can be identified with turnover times ranging up to many years. Considerations taken into account in developing biokinetic models for humans (see Section 3.3.6) are also relevant in developing biokinetic models for animals.

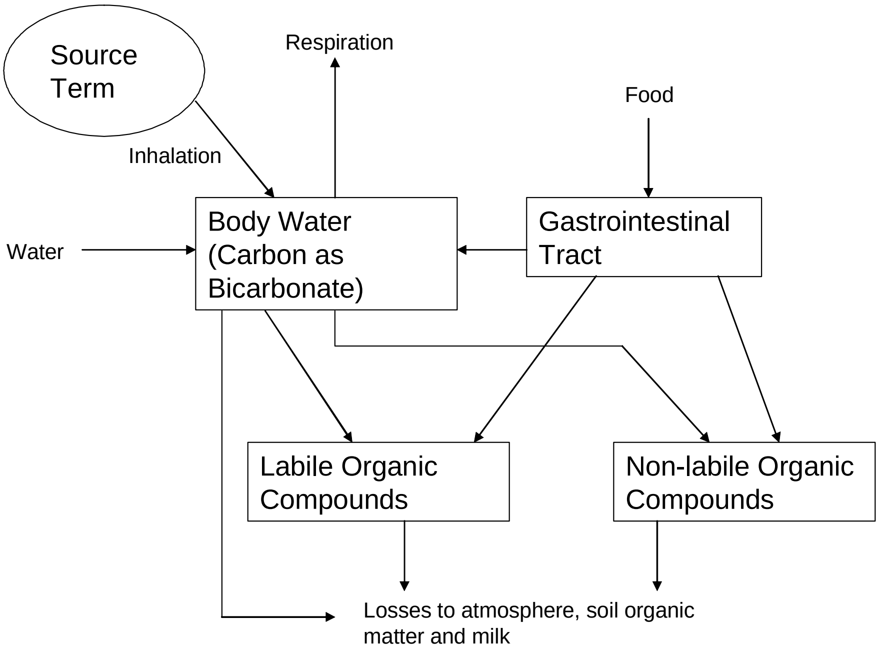

Figure 3.6: Model structure adopted for ^{3}H and ^{14}C in animals (Maul *et al.*, 2006).

3.3.6. Dose factors for ^{14}C with an emphasis on gaseous forms

Dose factors for ingested dietary ^{14}C and for various chemical forms of inhaled ^{14}C are available from the ICRP (1996). Dose factors for the various inhaled forms are listed in Table 3.3.

Table 3.3: Committed effective dose factors for various forms of ^{14}C (ICRP, 1996).

Chemical Form	Committed Effective Dose Coefficient (Sv Bq^{-1})					
	3 months	1 year	5 years	10 years	15 years	Adult
Aerosol (F)	6.1 10^{-10}	6.7 10^{-10}	3.6 10^{-10}	2.9 10^{-10}	1.9 10^{-10}	2.0 10^{-10}
Aerosol (M)	8.3 10^{-9}	6.6 10^{-9}	4.0 10^{-9}	2.8 10^{-9}	2.5 10^{-9}	2.0 10^{-9}
Aerosol (S)	1.9 10^{-8}	1.7 10^{-8}	1.1 10^{-8}	7.4 10^{-9}	6.4 10^{-9}	5.8 10^{-9}
Vapour	1.3 10^{-9}	1.6 10^{-9}	9.7 10^{-10}	7.9 10^{-10}	5.7 10^{-10}	5.8 10^{-10}
Carbon dioxide	1.9 10^{-11}	1.9 10^{-11}	1.1 10^{-11}	8.9 10^{-12}	6.3 10^{-12}	6.2 10^{-12}
Carbon monoxide	9.1 10^{-12}	5.7 10^{-12}	2.8 10^{-12}	1.7 10^{-12}	9.9 10^{-13}	8.0 10^{-13}

The aerosol forms listed are characterised by fast (F), medium (M) and slow (S) solubility related clearance from the respiratory system (ICRP, 1994). The vapour form is characterised by very fast uptake to the systemic circulation, with the ^{14}C subsequently being retained in the same way as dietary carbon.

Carbon dioxide and carbon monoxide give lower dose factors than the other forms considered because carbon dioxide is considered to be utilised to only a limited degree in metabolic processes and carbon monoxide is considered to bind only to haemoglobin and be lost to exhalation without other metabolic interactions (ICRP, 1981).

Although tritiated methane is covered in ICRP Publication 72 (ICRP, 1996), methane labelled with ^{14}C is not addressed. However, ICRP Publication 80 (ICRP, 1998), includes an addendum on ^{14}C-labelled methane. In that addendum, reference is made to the model for inhalation of ^{3}H-labelled methane described in ICRP Publication 71 (ICRP, 1995). In that model, methane is treated as an SR-1 vapour (soluble or reactive, see ICRP, 1994) with 1 % of the inhaled activity assumed to be absorbed in the lung and subsequently metabolised to form tritiated water.

This model was also applied to methane labelled with ^{11}C and ^{14}C. The available data indicated that the carbon label is oxidised to form carbon dioxide, but the possibility of some activity being incorporated into organic molecules was not excluded. The conservative assumption was made that one half of the metabolised fraction was retained with the half-life of carbon dioxide and one half with that of organic carbon. Under this assumption, the component retained like organic carbon dominates dosimetrically.

Using the model described above, the ICRP (1998) gave inhalation committed effective dose coefficients for ^{14}C-labelled methane of $6.6\ 10^{-12}$, $7.8\ 10^{-12}$, $4.9\ 10^{-12}$, $4.0\ 10^{-12}$, $2.9\ 10^{-12}$ and $2.9\ 10^{-12}$ Sv Bq^{-1} for ages of 3 months, 1 year, 5 years, 10 years, 15 years and adult, respectively.

The dosimetry of ^{14}C-labelled methane was investigated in detail by Phipps *et al.* (1990) and a summary of the results obtained was given in Nirex (1994). The dose factors from that summary are reproduced in Table 3.4.

Table 3.4: Effective dose rates from ^{14}C-labelled methane as used by Nirex (1994).

Age at Intake (y)	**Effective Dose Rate per Unit Concentration (Sv y^{-1} per Bq m^{-3})**			
	Absorbed Gas	**Gas in Lungs**	**Metabolised Gas**	**External Radiation**
1	$6.63\ 10^{-11}$	$2.49\ 10^{-10}$	$3.16\ 10^{-10}$	$2.20\ 10^{-10}$
10	$6.63\ 10^{-11}$	$1.74\ 10^{-10}$	$6.00\ 10^{-10}$	$2.20\ 10^{-10}$
20	$6.63\ 10^{-11}$	$9.15\ 10^{-11}$	$5.36\ 10^{-10}$	$8.84\ 10^{-11}$

Total effective dose rates were $8.51\ 10^{-10}$, $1.06\ 10^{-9}$ and $7.82\ 10^{-10}$ Sv y^{-1} per Bq m^{-3} for ages 1, 10 and 20 years, respectively.

These values can be converted into the same units as used in Table 2 by dividing by the appropriate breathing rates. These are 1900, 5300 and 6500 m^{3} y^{-1} for infants, children and adults (Nirex, 1997; Table 5.4). Thus, the committed effective dose factors are $4.5\ 10^{-13}$ Sv Bq^{-1} for infants, $2.00\ 10^{-13}$ Sv Bq^{-1} for children and $1.20\ 10^{-13}$ Sv Bq^{-1} for adults. These low values reflect the very limited uptake and metabolic utilisation of methane. In particular, they are an order of magnitude lower than the cautious values given by the ICRP (1998).

Subsequent to the studies reported above, little further work has been undertaken on the biokinetics and radiation dosimetry of ^{14}C-labelled gases. However, there is one further important paper on the biokinetics and dosimetry of ^{14}C-methane (Carlisle *et al.*, 2005).

Carlisle *et al.* (2005) pointed out that the ICRP value of 1% absorption is a conservative estimate, based on the observation that 0.33 % of infused ^{14}C-labelled methane was converted to ^{14}C-labelled carbon dioxide by sheep (Dougherty *et al.*, 1967). They noted also that these are the only published empirical data on the metabolism of radioactive methane. They also noted that the subsequent conversion of 1 % of the $^{14}CO_2$ to organically bound carbon, which is retained in the body with a biological half life of 40 days, was based on existing models derived from various human and animal experimental data.

Carlisle *et al.* (2005) undertook an experimental study in which rats inhaled ^{3}H-labelled or ^{14}C-labelled methane. They found that the ICRP model overestimated the uptake of radioactive methane in most tissues by an order of magnitude (values of 0.03 to 0.2 % were found for total ^{14}C retention one hour after a four hour exposure). When this effect had been taken into account, it was found that that the degree of conversion of retained ^{14}C to organically bound carbon was higher in some tissues than assumed by the ICRP, but that it was lower in others.

Based on their data, Carlisle *et al.* (2005) developed a dosimetric model for ^{14}C-labelled methane. This gave committed equivalent doses to the liver, carcass, brain and skin of 8.05 10^{-13} to 9.23 10^{-13}, 1.05 10^{-13}, 1.39 10^{-13} and 2.14 10^{-13} Sv Bq^{-1}, respectively. Overall, this suggests a committed effective dose per unit intake of about 1.5 10^{-13} Sv Bq^{-1}. This value is comparable with the values calculated by Phipps *et al.* (1990) and used in Nirex (1994).

Because the biokinetics and dosimetry of ^{14}C-labelled acetylene, ethylene and ethane had not been addressed in international recommendations or the peer-reviewed literature, Rees *et al.* (2003) developed a biokinetic and dosimetric model appropriate to ^{14}C-labelled ethylene. They argued that ethylene is a close chemical analogue of vinyl chloride monomer, differing from it only by the replacement of one hydrogen atom by a chlorine atom.

Vinyl chloride monomer is metabolised in the human body by the mixed-function oxidase system. The initial step is the formation of the epoxide, chloroethylene oxide , Figure 3.7.

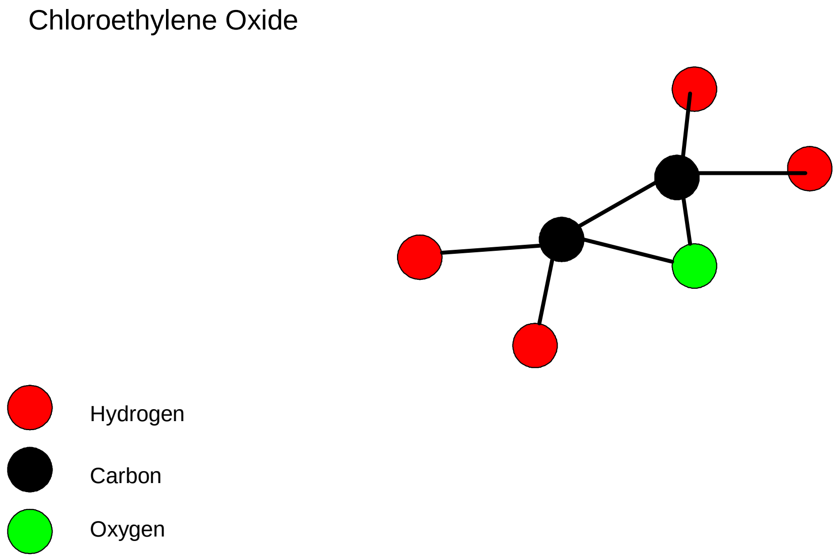

Figure 3.7: The structure of chloroethylene oxide.

Chloroethylene oxide can then spontaneously rearrange to form chloroacetaldehyde, which can be oxidised to chloroacetate. All of these compounds can bind directly, or by enzyme-mediated reactions, to glutathione. Hydroxylation reactions then give various cysteine compounds and hence thiodiglycollate, a urinary metabolite. Other secondary metabolic pathways are also thought to exist (Thorne *et al.*, 1986).

As discussed by Rees *et al.* (2003), ethylene is likely to be metabolised in a similar way. The double carbon bond could similarly be subject to epoxide formation by the mixed-function oxidase system, leading to a variety of metabolic products similar to those derived from vinyl chloride monomer.

Based on these considerations, Rees *et al.* (2003) adapted an existing biokinetic model for vinyl chloride monomer (Smith and Thorne, 1986) and applied it to ^{14}C-labelled ethylene in adults. Because of the high rate of turnover of metabolic products of ethylene, the committed effective dose factor obtained was 2.93 10^{-13} Sv Bq^{-1}, only slightly larger than that for ^{14}C-labelled methane.

Although there do not seem to be any directly relevant biokinetic data for ethylene, the epoxide, generally known as ethylene oxide, has been studied to a limited extent because of its significance as a toxic industrial chemical. The limited data that do exist confirm that it is efficiently metabolised, exhibits first-order kinetics and is rapidly excreted, but are not sufficient to inform development of a quantitative biokinetic model (ATSDR, 1990).

With respect to acetylene, it seems likely that the mixed function oxidase system would interact with the triple carbon bond, as this system is a general purpose system for the detoxification of a wide variety of substances and operates on substrates as diverse as benzene, vinyl chloride monomer, benzidine and its congeners, and carbon tetrachloride (Thorne *et al.*, 1986). It seems likely that products of such metabolism would be rapidly excreted in a way similar to those of vinyl chloride monomer.

For ethane, it seems plausible to assume that the degree of involvement in metabolic processes subsequent to inhalation would be similar to that of methane.

In assigning committed effective dose per unit intake values to ^{14}C-labelled acetylene, ethylene and ethane, it is reasonable to consider that the appropriate values should be much less than for ^{14}C-vapour (Table 3.3), which is assumed to be quantitatively incorporated in products of metabolism. Both methane and vinyl chloride monomer provide plausible biochemical analogues, and these yield comparable values of committed effective dose per unit intake, when the degree of conservatism incorporated in the ICRP (1998) model for methane is taken into account.

3.3.7. Aquatic systems

In general, ^{14}C entering freshwater systems in dissolved form can be treated as being dispersed in the same way as occurs for other radionuclides. Appropriate techniques for modelling such dispersion have been outlined by Thiessen *et al.* (1999). However, ^{14}C concentrations in aquatic organisms have often been calculated using a concentration ratio approach relative to ^{14}C concentrations in water (see, e.g. IAEA, 1994). This approach does not adequately address the fluxes of ^{14}C through aquatic foodchains. An ecosystem model of carbon cycling in a coastal area of the Baltic Sea has been developed for assessing the environmental transport and fate of a hypothetical discharge of ^{14}C over a period of 1000 years from the Swedish final repository for radioactive operational waste (SFR). The model involves identification, quantification and dynamic modelling of the main flows and storages of carbon, both in the physical environment as well as in the food web (Kumblad *et al.*, 2003). The processes included in the model were primary production, respiration, consumption, faeces production and water exchange. ^{14}C was in the model introduced into the food web via photosynthesising organisms and then transferred to higher trophic levels in proportion to the general carbon flow in the system and the ratio of ^{14}C to other carbon isotopes in the ecosystem components. Respired ^{14}C was connected to a compartment for dissolved inorganic carbon, which also was the inorganic carbon source for primary producing organisms. Thus, this mechanism provided re-circulation of respired carbon and ^{14}C in the system. A second pathway of re-circulation in the model was the excretion/loss of organic material by organisms that was connected to a compartment for particulate organic carbon (POM). POM was modelled to settle out to the sediment and become consumed by benthos and could thus become introduced in the food web again. The implications of changes of two parameters on the ^{14}C fate were examined: route of ^{14}C entry in the food web and water exchange rate. In general, the highest ^{14}C-concentrations were observed in benthic plants and benthic macro grazers, followed by fish and other organisms. ^{14}C entry into the food web via benthic primary producers was found to lead to increased concentrations in biota (especially benthic organisms) and reduced rates of water exchange were also observed to significantly increase the ^{14}C exposure of the organisms compared to a reference case.

This pioneering approach to ^{14}C modelling in the marine environment is now being extended from a compartmental approach to a high-resolution GIS-based approach (Wijnbladh *et al.*, 2006) and approaches based on carbon cycle models are also being developed for other types of environment (Ambio, 2006).

3.4. Effects of compartmental model structure and long-term inflow pollutograph on model predictions

Compartment models are often used in the radioecological modelling because they are simplistic conceptually, mathematically and easy to implement. They also have a realistic potential for handling the large number of relevant biogeochemical and ecological processes with low computing cost. However, care needs to be taken when selecting the number of compartments, and the basic assumptions limiting the potential conclusions need to be well understood. For an example, modelling of a tracer experiment is discussed in the following.

The field tracer experiment area is an artificial dam at Ekeby wetland in Sweden, Figure 3.8, with an area of 26 000 m^2 and a flow rate of 8 640 m^3 per day. Tritium was used as the tracer as a pulse of 74 GBq HTO injected within 5 h 15 min (Lindahl *et al.*, 2003).

Figure 3.8: Field tracer experiment at an artificial dam at Ekeby wetland, Sweden (Lindahl *et al.*, 2003).

The experimental residence time distribution was evaluated with an analytical solution based on compartment models with only one mass transfer rate between compartments known as “tanks-in-series” model, Figure 3.9. The basic assumption of instantaneous complete mixing in each tank prevails, as in any compartment model. This also implies that the spatial resolution of the results is limited to the scale of the tanks. The main feature with such a compartment structure is that the mass transfer rate governs both the advective and dispersive behaviour of the transport. The number of compartments is another variable that governs the transport behaviour.

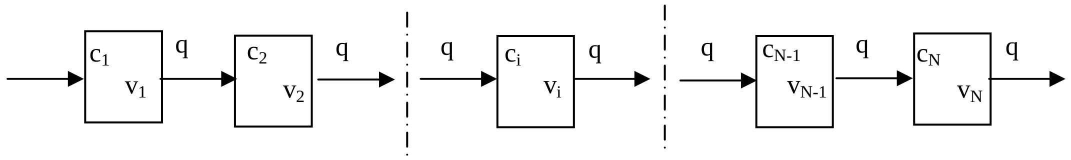

Figure 3.9: Conceptual “tanks-in-series” model (q = mass transfer rate, v_i = volume and c_i = concentration).

To explore the impact of model structure, a variable number of compartments was used and compared with the observations, Figure 3.10. When four compartments were used in the model, the simulated residence time distribution (RTD) could fit the measured data relatively well, except the fact that the compartment model was not able to describe a secondary peak caused by channelling in the wetland. It can be seen that when the input pulse is short, the model structure affects the output significantly.

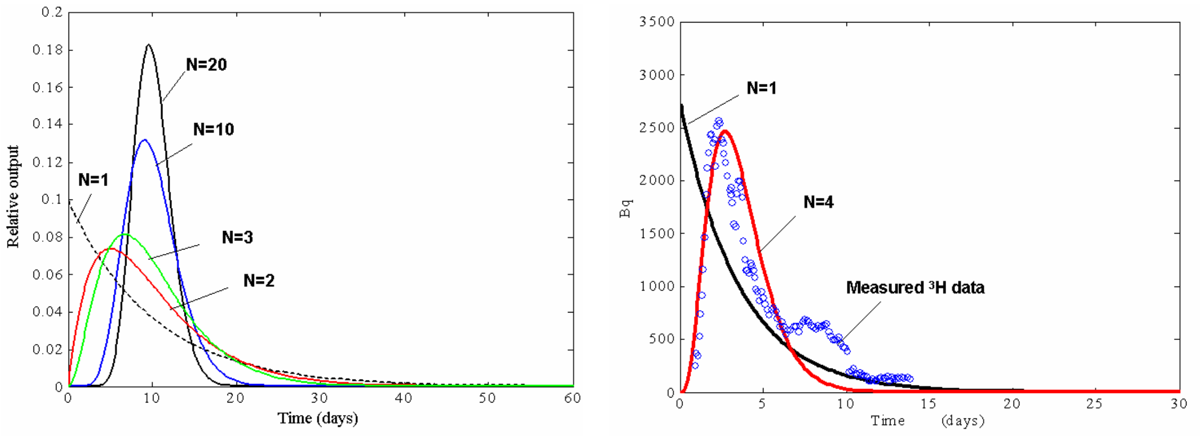

Figure 3.10: Response of a number of tanks to a Dirac delta pulse input in the “tanks-in-series” model (left) and modelled responses to an immediate pulse input of ^{3}H compared to the measured data (right).

The temporal moments of the residence time distribution are obtained from a general solution of the compartmental model in the Laplace domain derived with an arbitrary inflow pollutograph (Xu *et al.*, 2005). The obtained variance of the residence time, σ_t^2, can be used as a useful tool to analyse the effect of model structure and long-term inflow pollutograph on the response of the model predictions:

$$\sigma_t^2 = \sigma_t^2(0) + \frac{N}{q^2} = \sigma_t^2(0) + \sum_{i=1}^{N} \bar{t}_i^2 \qquad (3.5)$$

where *q* is the transport rate coefficient from compartment *i* to compartment *i+1* and also can be expressed as $q_i = 1/\bar{t}_i$ in which $\bar{t}_i$ is solute residence time in compartment *i*, *N* is the *N*th compartment at outlet, and $\sigma_t^2(0)$ is variance of residence time of the inflow pollutograph.

The effect of model structure and inflow pollutograph on the behaviour of the system can be analysed by means of equation (3.5). As can be seen from equation (3.5), the variance of residence time consists of two parts, one is from the temporal moments of the inflow pollutograph and the other from the temporal moments of the output of a unit pulse from the compartmental model. If the temporal moments related to the unit pulse through the compartmental model dominate this means that there could be a notable effect of model structure on the system response. A simple assumption made here is that residence time of solute transport in each compartment is proportional to the retardation factor. Equation (3.5) can thus be rewritten for a reactive solute as:

$$\sigma_t^2 = \sigma_t^2(0) + R^2 \frac{N}{q^2} = \sigma_t^2(0) + \sum_{i=1}^{N} R_i^2 \bar{t}_i^2 \qquad (3.6)$$

where *R* is retardation factor defined in Table 3.5 for various transport processes and $\bar{t}_i$ is the expected residence time for an inert solute in compartment i.

Assuming that the inflow pollutograph is a step injection type, the variance of inflow pollutograph can be expressed as:

$$\sigma_t^2(0) = \frac{\Delta^2}{12} \qquad (3.7)$$

where Δ is the duration of the inflow. From equations (3.6) and (3.7) it is clear that the influence of the model structure on the outflow pollutograph decreases as Δ tends towards infinity.

The example below illustrates how the effect of the model structure depends on Δ. Consider a case of transport of a medium-sorbing radionuclide, like caesium, in an aquatic system with a residence time of 14 days for inert water parcels (Xu *et al.*, 2005). Figure 3.11 shows the variance of residence time of the compartmental backflow model as a function of duration of inflow. As can be seen if the duration of the inflow pollutograph is shorter than ~10^2 years, the number of compartments has a significant effect on the variance of the residence time. As a conclusion, attention should be paid on selecting the number of the compartments, especially, for modelling time scales shorter that the characteristic time of the system to reach the steady state.

3.5. Modelling the input of radionuclides to the biosphere via groundwater

The development of radioactive waste disposal facilities requires implementation of measures that will afford protection of human health and the environment over a specific temporal frame that depends on the characteristics of the wastes. In order to demonstrate the satisfactory safety performance of any potential repository, performance assessments (PAs) have to be carried out (IAEA, 1995; Yim and Simonson, 2000). The repository design is based on a multi-barrier system: (i) the near field or engineered barriers, (ii) far field or geological barrier and (iii) the biosphere system. The characteristic reaction times and the dynamics of the biosphere are much more rapid than in the rest of the system. So, the coupling between the geosphere and biosphere processes is not necessarily simple. Here the focus is on the analysis of this frontier, the geosphere-biosphere interface zone (GBIZ). In reality, the transfer of radionuclides across the geosphere-biosphere interface can be very complex (Wasiolek *et al.*, 2005) due to several reasons.

Table 3.5: Retardation coefficients for different media (Xu *et al.*, 2005).

Medium	Retardation factor, R	Retardation coefficients
Bedrock	$1+2Z\frac{\varepsilon}{h}(1+\frac{\rho}{\varepsilon}k_d)$	$k_d = (c_w/c_m)\rvert_e$
Soil and lake ediments	$(1+K_D)$	$K_D = (c_p/c_d)\rvert_e$
Streams and rivers	$1+(1+K_B)\frac{\xi V_z PT}{2A}$	$K_B = \frac{\rho}{\varepsilon}k_d$

where Z = matrix "depth" in bedrock, ε = porosity, h = fracture aperture in bedrock, ρ = density, ξ = area reduction factor for the water exchange between a stream and the hyporheic zone, P = wetted parameter of a stream, V_z = exchange velocity for water between a stream and the hyporheic zone, T = residence time of inert solutes (water) in the hyporheic zone, A = cross-sectional area of stream, c_w = sorbed solute mass per unit solid mass, c_m = dissolved mass per unit volume of water, c_p = particulate solute mass per unit bulk volume, c_d = dissolved solute mass per unit bulk volume and $|_e$ denotes 'evaluated at equilibrium'.

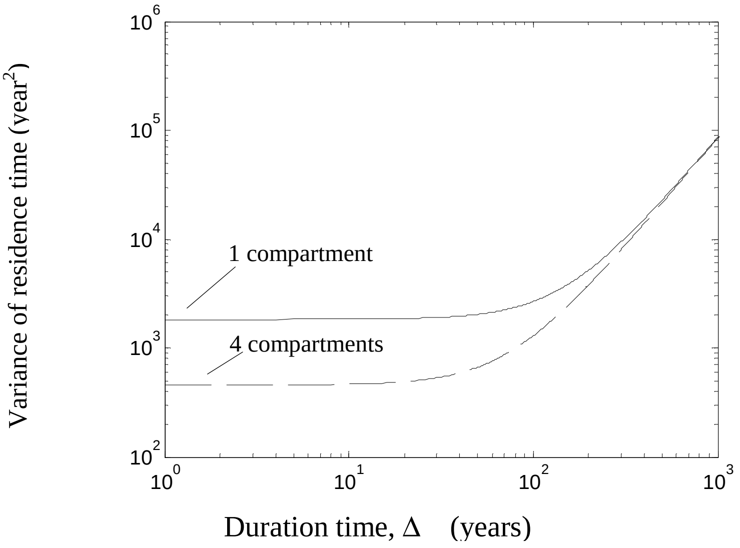

Figure 3.11: Comparison of the effect of a number of compartments based on variance of residence time versus inflow duration.

Based on recent data (see Wasiolek *et al.*, 2005) and on previous work published by Pinedo *et al.* (1999) findings on current approaches are summarised below:

- Well interfaces are common, but the treatment of near-surface groundwater flow and related radionuclide transport was not modelled very precisely in most of the cases reviewed. A key issue in the evaluation of this interface is where the well is situated relative to the distribution of contamination in the aquifer. Usually the peak concentration is assumed (pessimistic) or some average concentration over the whole aquifer (possibly optimistic) is considered. This difference in location can have a big effect on results.

- Dilution in the biosphere can be significant depending on the way in which the source term from the geosphere is defined. Examples of water flows at the GBIZ, depending on specific assumptions, range from 10^4 m^3y^{-1} for a minimal viable aquifer, 10^6 m^3y^{-1} for a small surface water body, 10^9 for a large surface water body or 10^{11} m^3y^{-1} for a marine release to a topographically constrained zone such as a bay or inlet.
- Direct discharge to the surface is considered in some PAs, especially for saturated media near to the surface. Release is either to a surface water body or to an area of subsoil. The assumption for volumetric flow in surface freshwaters is higher than the abstraction rates assumed for wells, ranging from about 10^6 to 10^9 m^3y^{-1}. For subsoil release, the area of soil contaminated is the most important parameter, determining output dose results.

The main modelling issues related to the geosphere-biosphere interface items which need to be explored in order to reduce uncertainties in the PA results are those related to: 1) the radionuclide source term from the geosphere to the biosphere, 2) the understanding and documentation of transport processes and parameters at the frontier and 3) the consideration of environmental and human induced changes due to the natural evolution.

3.5.1. Geosphere to biosphere radionuclide source term

The source term to the biosphere modelling needs to take account of the specific characteristics of the radionuclides involved, which have different physical, chemical and biological characteristics and behaviours and could require conceptually different models to predict their transport through, and distribution in, the relevant environmental media.

Only a limited number of radionuclides are likely to be important for the goundwater release and transport pathway from the point of view of their contribution to annual individual dose to humans: ^{237}Np (and its progeny), ^{129}I, ^{99}Tc, ^{36}Cl and ^{79}Se. For gaseous releases, ^{14}C and ^{36}Cl have to be considered. For early releases due to failure of the barrier system, consideration has to be given to ^{36}Cl, ^{79}Se, ^{107}Pd, ^{129}I, ^{99}Tc, ^{126}Sn, ^{135}Cs and U/Pu-series radionuclides including ^{237}Np. For human intrusion, inhalation pathways for Pu-, Am- and U-series radionuclides and external irradiation pathways for ^{94}Nb and ^{126}Sn could be relevant (Agüero *et al.*, 2006).

3.5.2. Understanding and documentation of main processes and parameters

The importance of individual processes controlling radionuclide transfer to the biosphere is dependent on the characteristics of the site, in terms of the geology, hydrogeology, topography, soil system, climate and vegetation (Hooker *et al.*, 2002). As such, radionuclide transfer processes are site and system specific. The movement of surface water and groundwater is controlled to a large extent by the topography and geologic characteristics of an area. In addition, climate, through the effects of precipitation and evapo-transpiration, controls the delivery of water to and from the Earth´s surface. Therefore, it is necessary to understand the effects of topography, geology, and climate on surface-water and groundwater flow systems in order to understand the interactions of groundwater and surface waters potentially contaminated with radionuclides. The following specific issues are identified as requiring consideration.

- Further work could be done to take better account of the effects of meteoric waters mixing with contaminated deeper water discharging into the near surface (Wasiolek *et al.*, 2005), besides analyses of the process of capillary rise of contaminated water through the unsaturated zone.
- In addition, further consideration might be given to hydro-geochemical processes that could affect radionuclide migration in the near surface, resulting in accumulations which could then be released as a result of environmental change (Wasiolek *et al.*, 2005). Coupling between processes, in particular couplings to geochemistry are regarded as a direction to follow. A variety of 1D, 2D and 3D mathematical models

exist to represent these processes in combination. Approaches such as particle tracking in 2D and 3D fracture-network or equivalent porous medium representations can now give useful indications of release locations and areas. In dry areas, direct discharge to the surface would dry out, resulting in the potential for high accumulation. Flow into the marine environment results in high dilution and much lower doses than the equivalent discharge to a terrestrial or lacustrine environment. For some sites, sea-level change could result in terrestrial releases becoming marine, and *vice versa* (Wasiolek *et al.*, 2005).

- Furthermore, the impact of water abstraction volumes and rates on the groundwater model domain must be considered.
- A necessity to explore the significance of natural accumulation pathways in the biosphere has been identified (Klos and Albrecht, 2005; Simón *et al.*, 2005), so it seems to be relevant to explore these matters also in the context of the GBIZ.
- An analysis of redox effects on soil distribution coefficient values and how to make partitions of the soil profile into oxygenated and non-oxygenated layers for modelling purposes is required (Agüero *et al.*, 2006).
- Sensitivity analyses can be used to check whether there are conditions for which a particular process should be explicitly included in a PA model.
- Appropriate documentation of Features, Events and Processes (FEPs) and development of conceptual models through the use of the Interaction Matrix (IM) approach (Hudson, 1992) is recommended. The IM has proved powerful in ensuring that all relevant processes are taken into account in specifying a conceptual model of radionuclide transport in the biosphere and this approach could help to support the reduction of uncertainties in the PA approach relating to the GBIZ (Agüero *et al.*, 2005).
- Analysis of source-pathway-receptor for scenario development is suggested, because this has the power of combining forward (source-pathway) and backward (receptor-pathway) analyses to address both how could someone could be exposed and how a release across a GBIZ could lead to exposure (Wasiolek *et al.*, 2005).

3.5.3. Environmental Change

The long periods of time of relevance in PA and the inherent difficulty in projecting the precise biosphere conditions that will arise make it impossible to validate, strictly speaking, models of the overall behaviour of the system (although validation of the representation of some processes or component submodels is possible) (Whicker *et al.*, 1999; Agüero, 2005). In addition, over the long time scales considered, modifications of the GBIZ are certain to occur, due to both natural and man-made factors, and, in particular, climatic and geomorphological changes. Specific matters that need to be considered include the following.

- The limitations of the current scientific understanding of the climate system need to be clearly stated when the results of climate modelling are used in support of PA (Texier *et al.*, 2003). A better integration of basic climatology, geography and ecology into models of the evolution of the environment as it affects radionuclide transport at the GBIZ would be desirable.
- Due to the difficulties to predict the human activities in the long term, or even beyond a few years into the future, the international consensus and recommendation is to use present-day data for areas with similar environmental characteristics to the one under analysis.
- In addition more studies are needed in order to quantify accurately environmental change processes and rates in order to obtain parameter values for modelling purposes.

4. Needs for research and development for each radionuclide

In this section the suggestions of the group on needs for research and development for each specific radionuclide are summarised.

4.1. ^{14}C

The following issues were be considered by the group as priorities for research concerning ^{14}C behaviour in terrestrial ecosystems.

- Metabolism of methane in soil and the interaction between the soil solution and the soil atmosphere.
- Root uptake by plants to ascertain if this is a process of importance or not.
- Uptake from the sub-canopy and above-canopy atmosphere.
- Losses from irrigation waters during extraction and application.
- Degree of uptake of ^{14}C from ingested food in the gastrointestinal tract.
- Characterisation of body pools due to difference in metabolic rates among tissues and organs.

The following drivers should be considered:

- land use;
- climatic conditions: moist, temperature, pressure;
- soil and sediment chemistry;
- hydrology.

For long-term understanding of the time evolution of ^{14}C in the environment, an analysis of scenarios that could lead to changes in the system should be performed in accordance with data already obtained by the international climate change panel (ICCP). This is important for the long-term protection of the environment.

The group considers that the development of a system for classification of ecosystems based on their relative importance in global carbon cycle should be envisioned. The description of the processes represented in the matrices for terrestrial and aquatic ecosystems and the implications for ^{14}C modelling should be reinforced. More importantly, the factors controlling the processes should be clearly defined and should include interactions between ecosystems. The proposed matrices have to be considered as non-exhaustive. Therefore, the group recommends further work on these matrices in order to achieve an as complete as possible description of all the potential processes governing the behaviour of ^{14}C in the environment.

One important task concerning modelling of ^{14}C is the identification of available radioecological and ecological models. The group is critical of the use of CF, in its classical definition, to describe ^{14}C transfer and recommends alternatives, such as the specific activity approach and dynamic models. The group noted that there are good models of carbon cycling in all types of ecosystem, which could be adapted to the problem of long-term ^{14}C cycling. The selection of the appropriate model is a matter of defining the appropriate temporal and spatial scales for the problem. For long time scales, consideration of the effects of climate change will be necessary, especially if there is an interest in protection of the environment.

4.2. ^{36}Cl

The group recommended that the following priority areas for research and development should be taken into account for improving the understanding of the long-term behaviour of ^{36}Cl in the environment:

- More experiments for the determination of ^{36}Cl soil-to-plant transfer should be performed and combined with studies of stable Cl behaviour. In particular, direct measurement of uptake chlorine fluxes by forest vegetation and reliable estimations of budgets are needed. In general, there is a need for measurements of the Cl content in natural vegetation, It is also necessary to consolidate data on the rates of ^{36}Cl translocation from wet or dry deposits to plants. The origin and dynamics of Cl retained by forests and other semi-natural ecosystems is unclear. For elucidating this, it is necessary to improve the quantitative description of Cl recycling in the soil-subsoil system. Long term effect of changes in boundary conditions on Cl budget should also be considered.
- The role of organic Cl (Cl_{org}) in the cycling of this element cannot be ignored, but quantitative estimates of Cl_{org}/Cl_{inorg} in plants and soils are still scarce. Hence, specific measurements of organic and inorganic chlorine should be performed in future studies. The chlorination/mineralisation rates of organic matter in different climatic and ecological conditions need to be determined. Indeed, this may lead to determination that the high mobility of chlorine may be impaired in its organic forms. Organic Cl volatilisation processes should also be studied.

Concerning modelling of ^{36}Cl long-term behaviour in the environment, the group proposes to compare existing and currently under development models of different types:

- specific activity models;
- kinetic models;
- equilibrium models;
- combinations of the above types.

This model comparison should allow the scientific issues associated with ^{36}Cl behaviour to be addressed, but must be structured to take into consideration radiological endpoints, bearing in mind that the importance of dynamic factors will be partly determined by what is meant by the average dose to a critical group.

The group considered that the balance of the input and output of stable chlorine into the system is an important factor that needs to be taken into account when modelling ^{36}Cl migration in the environment.

The group also proposes inclusion, as far as possible, in the new models the processes effectively describing the role of organic ^{36}Cl in the different environmental compartments and more specifically in the soil/plant subsystem. This means that the currently proposed matrices (see Annex) are not comprehensive and enhancements will be required in the future.

Finally, validation of the already existing models and of those currently under development necessitates sets of data concerning both organic and inorganic ^{36}Cl. In that context, the needs of modellers are complementary to those of experimentalists.

4.3. ^{99}Tc

There is a need to develop mechanistic models of the speciation of technetium in the environment and of the relationship between speciation and bioaccumulation. Limited data to provide parameter values needed for modelling purposes in terrestrial environments suggest performing experimental work on:

- Obtaining distribution coefficient values (K_d) covering different redox conditions, including scenarios for reoxidation of Tc sorbed in the soil and the impact of climate changes on the mobility of the radionuclide.
- Obtaining data of soil-to-plant concentration factors (CF), especially for non-agronomic, natural, plants.
- Studying the behaviour of Tc in humic systems, which have been poorly studied till now.

4.4. ^{237}Np

In the case of neptunium the group considered that there is a need for performing reviews of the literature with a focus on key processes, like transfers to plants and animals, and the accumulation and bioavailability of the neptunium in soil. Also, valuable information about neptunium metabolism in man could be obtained from the database on workers' exposure to transuranic elements existing in the US Registry of Workers exposed to Transuranic Elements.

The following priorities for experimental research were identified:

- To analyse the database of information on workers' exposure to transuranic elements existing in the US Registry of Workers exposed to Transuranic Elements.
- Experimental studies of metabolism of Npin large animals are required. Bioavailability studies could be carried out using ^{239}Np.
- It is recommended that analyses of Np in samples of soils and plants are carried out, for estimating soil-to-plant transfer factors, using more advanced analytical techniques that have been developed during recent years. For the analyses, samples contaminated with Np from fallout that exist in storage at various laboratories could be used.
- Further development of analytical methods, for example neutron activation techniques, is required for performing measurements of important parameters such as *in situ* sorption in saline and fresh waters.
- It is necessary to carry out food chain studies, such as the lichen-reindeer-man food chain, using available samples and applying newly developed techniques.
- Studies of Np behaviour in anoxic fjords and meromictic lakes are required.
- There is a need to carry out research for improving the understanding of the effects of redox conditions on Np solubility and mobility in the environment.

4.5. ^{238}U

4.5.1. Terrestrial environments

Geochemical modeling should be more used in soil-plant transfer studies. There is a good knowledge of the U geochemistry, which is well integrated in speciation predictive tools. This is an advantage for modeling of uranium adsorption on mineral assemblages and of uranium solubility in soils. That knowledge is relevant also to assist in developing strategies for remediation of contaminated sites, where long-term effects of various amendments and vegetation development should be considered.

There is still a large range of pH values ($5<pH<8$) where the effect of soil-plant interactions on U availability and uptake is poorly understood. The influence of the following rhizospheric processes is possibly of importance:

- major element depletion or enrichment (e.g. P, Ca, "iron" plaque);
- acidification/alkalinisation;

- organic exudates and further ligand exchange.

The existing K_d and TF values have been mainly derived from empirical studies using simplistic approaches. It is necessary to enlarge the past focus on “transfer” to more mechanistic experimental approaches and modeling. That involves the identification/quantification of:

- mechanisms of U absorption and sequestration in roots linked to the presence or absence of myccorrhizae and bacteria;
- speciation and "inactivation" in various organs, at cellular and subcellular level and in xylem sap;
- influence of transpiration and uptake mechanisms: passive-active;
- effect of plant nutrient deficiency and organ senescence on U accumulation and translocation;
- uranium behaviour in terrestrial environments, which is characterised by a large range of K_d and TF values. It is desirable to explore a better categorisation of U mobility and transfer coefficients in terms of intrinsic soil and plant properties with emphasis on root external-root internal-shoot (fruit) translocation,

In addition, a more integrated approach in field studies is required, with emphasis on exchange dynamics between compartments (flux calculations) and interactions between water, biomass and element cycling in contrasting ecosystems.

4.5.2. Aquatic environments

Knowledge gaps remain in several aspects of the uranium cycle in aquatic systems, such as the role of sediments as reservoirs, the role of colloids, the exchange processes between solution and suspended particles, the role of microbiology and organic matter. It is also necessary to better understand the relationship between chemical speciation, bioavailability and bioaccumulation.

The use of “equilibrium” assumptions (e.g. based on mean K_d and TF values) would necessarily limit the predictive ability of the transfer models, without any possible generalisation to situations where kinetics of reactions and biological processes dominate. The introduction of chemical models including kinetic reactions would be potentially powerful.

Another aspect is the linking between chemistry (chemical speciation) and biology (bioavailability and biokinetics). Existing models for other trace metals such as FIAM and BLM (Free-Ion Activity Model; Biotic Ligand Models – see Campbell (1995); Slaveykova and Wilkinson (2005) - for a comprehensive overview) have already shown their potential power in predicting uranium bioavailability in contrasted environments (Markich *et al.*, 1996; Fortin *et al.*, 2004; Denison, 2004). FIAM/BLM models are however insufficient to fully explain the mechanisms involved in U bioavailability to freshwater organisms (algae, bivalves, fish). Biological transport kinetics may depend on the same parameters that influence speciation and competition for sites due to acid-base and ion regulation. Considering, in addition, that the thermodynamic data used to model the speciation of uranium are insufficiently precise, a major consequence is the current difficulty in distinguishing between abiotic and biotic factors (Denison, 2004; Fortin *et al.*, 2004).

5. General conclusions and recommendations

This report provides an overview of the available knowledge related the behaviour of ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np, ^{238}U in both terrestrial and aquatic ecosystems based on information that has been collated by experts in the field, who participated in three scientific workshops organised by the IUR Task Force "Radioecology and Waste". The findings have been made available to the BIOPROTA group, and collaboration with BIOPROTA has been maintained throughout the work of the Task Force.

Interaction matrices have been developed for terrestrial and aquatic environments. These matrices identify the main components and processes that describe the behaviour of the radionuclides in these two broad environment types. Furthermore, the potential relevant processes were identified for each specific studied radionuclide (^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np, ^{238}U). The matrices are not fully comprehensive and should be improved in terms of completeness of the processes involved and the identification of the key processes for each radionuclide.

For each studied radionuclide, the most important processes that need to be considered in long-term assessments were identified and recommendations are given for these processes on research needed for filling in data and knowledge gaps. There is a wide difference in data and process descriptions between the five studied radionuclides. For ^{14}C there is a good understanding of the behaviour, supported by empirical data, as opposed to ^{237}Np where there are few empirical data and assessments are usually done using data and knowledge for analogues, *e.g.* uranium and plutonium. New experimental studies should therefore be initiated to fill these gaps.

In the future work of the Task Force, it is necessary to describe the important processes in more detail and propose ways of modelling. Possible approaches for improving the models have been outlined in this report. For ^{14}C a detailed model is described and can be used.

The group recommends that the Task Force should apply the interaction matrices to other radionuclides, supported by descriptions of the relevant processes. Preferred ways of modelling for these new radionuclides should also be identified. The following list of radionuclides to be considered is proposed: ^{59}Ni, ^{79}Se, ^{94}Nb, ^{129}I, ^{226}Ra, ^{239}Pu, and ^{241}Am.

The developed interaction matrices are detailed and would lead to complex models if all components were modelled. By identifying the most important processes, it should be possible to reduce the complexity of the models. However, it must be remembered that justification must be given in order to move from a detailed model to a simplified model that is fit for the assessment purposes.

The IUR "Radioecology and Waste" Task Force has met once a year, inviting specialists to discuss specific issues. This approach has been proven to work well and it is proposed that the Task Force continues to work in the same way and at the same level of participation. The use of interaction matrices has also been successful within the group in facilitating discussions and documenting the views of the experts.

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Appendix 1: Terrestrial matrices for individual radionuclides: ^{14}C, ^{237}Np, ^{36}Cl and ^{238}U

From the general matrix for terrestrial environments in Figure 2.1 of Section 2.1, an explicit matrix was developed for ^{14}C, ^{237}Np, ^{36}Cl and ^{238}U. ***Application to ^{14}C* –** This terrestrial matrix shows the processes of potential importance for ^{14}C highlighted in bold.

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)
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
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)Exhation **2) Erucaltation**	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)
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)
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
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
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
1)Resuspension (at outcrop)	1)Desorption 2)Mineral dissolution	1)External contamination	1)Ingestion 2)Bioturbation	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

Application to ^{36}Cl - This matrix shows the general interaction matrix for the terrestrial environment, with the processes of potential importance for ^{36}Cl highlighted in bold.

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)
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**
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) Eructation	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)
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)
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
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
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
1)Resuspension (at outcrop)	1)Desorption 2)Mineral dissolution	1)External contamination	1)Ingestion 2)Bioturbation	**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

Application to ^{237}Np - This matrix shows the general interaction matrix for the terrestrial environment, with some processes of potential importance for ^{237}Np highlighted in bold.

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)
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
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) Eructation	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)
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)
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
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
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
1)Resuspension (at outcrop)	1)Desorption 2)Mineral dissolution	1)External contamination	1)Ingestion 2)Bioturbation	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

Application to ^{238}U - This matrix shows the general interaction matrix for the terrestrial environment, with the processes of potential importance for ^{238}U highlighted in bold.

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)
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**
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) Eructation	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)
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)
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**
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
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**
1)Resuspension (at outcrop)	**1)Desorption** **2)Mineral dissolution**	**1)External contamination** **2)Irrigation**	1)Ingestion 2)Bioturbation	**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 2: Aquatic matrices for individual radionuclides: ^{14}C and ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U

From the general matrix in Figure 2.2 of Section 2.3, a non-exhaustive matrix was developed for ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U.
Application to ^{14}C - This matrix shows the general interaction matrix for the aquatic environment, with the processes of potential importance for ^{14}C highlighted in bold.

Atmosphere	**1)Deposition**							1)Diffusive exchange 2)Pressure pumping
1)Evaporation **2)Gas evolution** 3)Droplet production	Water	Adsorption, complexation, **precipitation**	1)Diffusion 2)Advection 3)Adsorption 4)Complexation 5)Precipitation	1)Uptake **2)photosynthesis** 3)deposition	1)Uptake	1)Uptake	**1)Uptake**	1)Recharge by surface waters
1)Resuspension	1)Desorption **2)Dissolution**	Abiotic Suspended Matter	1)Deposition		1)Ingestion	1)Ingestion	1)Ingestion 2)Uptake	1)Particle transport 2) Colloidal transport
1)Resuspension	1)Desorption **2)Dissolution** 3)Degassing	1)Erosion of bed sediment	Deposited Matter (sediment)	1)Root uptake	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Particle transport 2) Colloidal transport
1)Respiration 2)Release of other organic matter	**1)Respiration** **2)Release of other organic matter**		**1)Death**	Primary Producers (phytoplankton, macrophytes, aquatic plants)	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Biological weathering
1)Exhalation 2)Eructation	**1)Exhalation** 2)Eructation **3)Excretion**		**1)Death** 2)Bioturbation		Consumers I (zooplankton, macrobenthos)	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation **3)Excretion**		**1)Death** 2)Bioturbation			Consumers II (omnivorous fish)	1)Ingestion 2)Uptake	1)Excretion 2)Death and decomposition
1)Resuspension	1)Resuspension **2)Excretion**		1)Death 2)Bioturbation		**Grazing/Uptake**		Decomposers	1)Excretion 2)Death and decomposition
1)Resuspension	1)Desorption 2)Mineral dissolution	1)Chemical mechanical weathering	1)Chemical mechanical weathering		1)Ingestion 2)Bioturbation	1)Ingestion 2)Bioturbation	1)Ingestion 2)Utilisation	Interface with Geosphere

Application to ^{36}Cl - This matrix shows the general interaction matrix for the aquatic environment, with the processes of potential importance for ^{36}Cl highlighted in bold.

Atmosphere	**1)Deposition**							1)Diffusive exchange 2)Pressure pumping
1)Evaporation **2)Gas evolution** 3)Droplet production	Water	Adsorption, complexation, precipitation	1)Diffusion 2)Advection 3)Adsorption 4)Complexation 5)Precipitation	**1)Uptake** 2)photosynthesis 3)deposition	**1)Uptake**	**1)Uptake**	**1)Uptake**	1)Recharge by surface waters
1)Resuspension	1)Desorption 2)Dissolution	Abiotic Suspended Matter	1)Deposition		1)Ingestion	1)Ingestion	1)Ingestion 2)Uptake	1)Particle transport 2) Colloidal transport
1)Resuspension	1)Desorption 2)Dissolution 3)Degassing	1)Erosion of bed sediment	Deposited Matter (sediment)	1)Root uptake	1)Ingestion	1)Ingestion	1)Ingestion **2)Uptake**	**1)Particle transport** 2) Colloidal transport
1)Respiration 2)Release of other organic matter	1)Respiration 2)Release of other organic matter		**1)Death**	Primary Producers (phytoplankton, macrophytes, aquatic plants)	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Biological weathering
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		**1)Death** 2)Bioturbation		Consumers I (zooplankton, macrobenthos)	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		**1)Death** 2)Bioturbation			Consumers II (omnivorous fish)	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Resuspension	**1)Resuspension** **2)Excretion**		**1)Death** 2)Bioturbation		Grazing/Uptake		Decomposers	1)Excretion 2)Death and decomposition
1)Resuspension	1)Desorption **2)Mineral dissolution**	1)Chemical mechanical weathering	1)Chemical mechanical weathering		1)Ingestion 2)Bioturbation	1)Ingestion 2)Bioturbation	1)Ingestion 2)Utilisation	Interface with Geosphere

Application to ^{99}Tc - This matrix shows the general interaction matrix for the aquatic environment, with the processes of potential importance for ^{99}Tc highlighted in bold.

Atmosphere	1)Deposition							1)Diffusive exchange 2)Pressure pumping
1)Evaporation 2)Gas evolution 3)Droplet production	Water	**Adsorption, complexation, precipitation**	**1)Diffusion** **2)Advection** **3)Adsorption** **4)Complexation** **5)Precipitation**	**1)Uptake** **2)photosynthesis** **3)deposition**	**1)Uptake**	**1)Uptake**	**1)Uptake**	1)Recharge by surface waters
1)Resuspension	**1)Desorption** **2)Dissolution**	Abiotic Suspended Matter	**1)Deposition**		**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Particle transport 2) Colloidal transport
1)Resuspension	**1)Desorption** **2)Dissolution** 3)Degassing	**1)Erosion of bed sediment**	Deposited Matter (sediment)	**1)Root uptake**	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Particle transport 2) Colloidal transport
1)Respiration 2)Release of other organic matter	1)Respiration **2)Release of other organic matter**		**1)Death**	Primary Producers (phytoplankton, macrophytes, aquatic plants)	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Biological weathering
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation **3)Excretion**		**1)Death** 2)Bioturbation		Consumers I (zooplankton, macrobenthos)	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation **3)Excretion**		**1)Death** 2)Bioturbation			Consumers II (omnivorous fish)	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Resuspension	1)Resuspension **2)Excretion**		**1)Death** 2)Bioturbation		**1)Ingestion** **2)Uptake**		Decomposers	1)Excretion 2)Death and decomposition
1)Resuspension	1)Desorption 2)Mineral dissolution	1)Chemical mechanical weathering	1)Chemical mechanical weathering		1)Ingestion 2)Bioturbation	1)Ingestion 2)Bioturbation	1)Ingestion 2)Utilisation	Interface with Geosphere

Application to ^{237}Np and ^{238}U - This matrix shows the general interaction matrix for the aquatic environment, with the processes of potential importance for ^{237}Np and ^{238}U highlighted in bold.

Atmosphere	1)Deposition							1)Diffusive exchange 2)Pressure pumping
1)Evaporation 2)Gas evolution 3)Droplet production	Water	**Adsorption, complexation, precipitation**	**1)Diffusion** **2)Advection** **3)Adsorption** **4)Complexation** **5)Precipitation**	**1)Uptake** 2)photosynthesis 3)deposition	**1)Uptake**	**1)Uptake**	**1)Uptake**	1)Recharge by surface waters
1)Resuspension	**1)Desorption** **2)Dissolution**	Abiotic Suspended Matter	**1)Deposition**		**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Particle transport 2) Colloidal transport
1)Resuspension	1)Desorption 2)Dissolution 3)Degassing	1)Erosion of bed sediment	Deposited Matter (sediment)	**1)Root uptake**	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Particle transport 2) Colloidal transport
1)Respiration 2)Release of other organic matter	1)Respiration 2)Release of other organic matter		**1)Death**	Primary producers (phytoplankton, macrophytes, aquatic plants)	**1)Ingestion**	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Biological weathering
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		**1)Death** 2)Bioturbation		Consumers I (zooplankton, macrobenthos)	**1)Ingestion**	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Exhalation 2)Eructation	1)Exhalation 2)Eructation 3)Excretion		**1)Death** 2)Bioturbation			Consumers II (omnivorous fish)	1)Ingestion **2)Uptake**	1)Excretion 2)Death and decomposition
1)Resuspension	1)Resuspension 2)Excretion		**1)Death** 2)Bioturbation		**1)Ingestion** **2)Uptake**		Decomposers	1)Excretion 2)Death and decomposition
1)Resuspension	1)Desorption **2)Mineral dissolution**	**1)Chemical mechanical weathering**	**1)Chemical mechanical weathering**		1)Ingestion 2)Bioturbation	1)Ingestion 2)Bioturbation	1)Ingestion 2)Utilisation	Interface with Geosphere

Appendix 3: Members of the IUR "Radioecology and Waste" Task Force and workshop participants

Members of the IUR Radioecology and Waste Task Force since 2002

Name	Organisation	Country	Participation to IUR "Radioecology and Waste" workshops			
			Monaco, ***4th Sept'02***	**Merlewood,** ***27-28 Feb'03***	**Madrid,** ***3-4 Nov'03***	**Mol,** **18 - 19 Oct'06**
Rodolfo Avila - Task Force Leader	Facilia AB	Sweden	y	y	y	y
Achim Albrecht	ANDRA	France				
Adrian Punt	Enviros	UK			y	
Almudena Agüero	CIEMAT	Spain		y	y	y
Anders Wörman	SLU	Sweden				
Andre Maes	K.U.Leuven	Belgium				
Ari Ikonen	Posiva	Finland			y	y
Brenda Howard	CEH	UK	y	y	y	
Carine Damois	EdF	France		y		
Carl-Magnus Larsson	SSI	Sweden	y			
Christian Tamponnet	IRSN	France		y		y
Claus Bunnenberg	ZSR, University of Hannover	Germany	y			
Cyril Feidt	ENSAIA-INPL	France				
Dan Galeriu	Inst. Physics	Romania		y	y	
Daniel Ashworth	Imperial College	UK		y	y	
Duncan Jackson	Enviros	UK		y		

Name	**Organisation**	**Country**	**Participation to IUR “Radioecology and Waste” workshops**			
			Monaco, ***4th Sept’02***	**Merlewood,** ***27-28 Feb’03***	**Madrid,** ***3-4 Nov’03***	**Mol,** **18 - 19 Oct’06**
Elis Holm	Riso/Lund	Sweden		y	y	
Elisabeth Leclerc-Cessac	ANDRA	France		y		
Frits van Dorp	Nagra	Switzerland				
George Shaw	Imperial College	UK				
Graham Smith	Enviros	UK			y	
Guillaume Echevarria	ENSAIA-INPL	France				
Helene Stensrud	NRPA	Norway	y			
Hildegarde Vandenhove	SCK-CEN	Belgium	y	y		
Inmaculada Simón	CIEMAT	Spain			y	
Irene Zinger	Facilia AB	Sweden				y
Ives Thiry	SCK-CEN	Belgium	y		y	y
Jane Clifford	CEH	UK				
Jean Claude Barescut	IRSN	France				
Kath Morris	University of Leeds	UK		y		
Linda Kumblad	SKB	Sweden		y		
Lionel Dewiere	IRSN	France	y			
Marsha Sheppard	Ecomatters	Canada				
Mike Thorne	MTA Ltd	UK		y		
Nick Beresford	CEH	UK		y		
Nick Ostle	CEH	UK		y		

Name	Organisation	Country	Participation to IUR “Radioecology and Waste” workshops			
			Monaco, *4th Sept’02*	Merlewood, *27-28 Feb’03*	Madrid, *3-4 Nov’03*	Mol, 18 - 19 Oct’06
Philippe Guetat	CEA	France	y			
Rodolphe Gilbin	IRSN	France			y	y
Ronny Bergman	FOI	Sweden	y			
Shulan Xu	SSI	Sweden			y	
Stamatia Massoura	(LSE)-ENSAIA	France		y		
Steve Sheppard	Ecomatters	Canada				
Ulrik Kautsky	SKB	Sweden	y		y	
Umberto Sansone	ANPA	Italy				
Valery Kashparov	UIAR	Ukraine		y		
Xiaolin Hou	Riso/Lund	China		y		

Participants in the Workshops

Merlewood - 27-28 February 2003

D. Ashworth D. Galeriu D. Jackson
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Mol - *18 – 19 October 2006*

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Appendix 4: Organisation Abbreviations

IUR	International Union of Radioecology
(LSE)-ENSAIA	Laboratoire Sols et Environnement, France
ANDRA	Agence Nationale pour la Gestion des Déchets Radioactifs
ANPA	The Agency for Environmental Protection and Technical Services - Associazione Nazionale Praticanti ed Avvocati
CEA	Commissariat à l'Energie Atomique
CEH	Centre for Ecology and Hydrology, UK
CIEMAT	Research Centre in Energy, Environment and Technology
EdF	Electricité de France
ENSAIA-INPL	Laboratoire de Science et Génie Alimentaires, France
FOI	Swedish Defence Research Agency
ICSTM	Imperial College of Science, Technology and Medicine (University of London)
IRSN	Institute for Radiological Protection and Nuclear Safety
K.U.Leuven	Katholieke Universiteit Leuven, Belgium
MTA Ltd	Mike Thorne and Associates Ltd
Nagra	National Cooperative for the Disposal of Radioactive Waste
NRPA	Norwegian Radiation Protection Authority
SCK-CEN	Belgian Nuclear Research Centre
SKB	Swedish Nuclear Fuel and Waste Management Company
SLU	Swedish University of Agricultural Sciences
SSI	Swedish Radiation Protection Authority
UIAR	Ukranian Institute of Agricultural radiology
ZSR, U. Hannover	Center fro Radiation Protection and Radioecology, University of Hannover, Germany

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