$\mathcal{B}$IOPROTA

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

THEME 2: Task 3:

Model Review and Comparison for C-14 Dose Assessment

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APPENDIX A. SPECIFIC ACTIVITY MODELS IN RELATION TO BIOTRAC BIOSPHERE MODEL.

APPENDIX B. THE IMPLICATIONS OF RELEASES OF C-14 IN GASEOUS FORM FROM A RADIOACTIVE WASTE REPOSITORY, BY M C THORNE

FOREWORD

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

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

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

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

BIOPROTA Concept

BIOPROTA provides a forum to address uncertainties in the assessment of the radiological impact of releases of long-lived radionuclides into the biosphere. The programme of work carried out under the auspices of BIOPROTA focuses on these key radionuclides and the various biosphere migration and accumulation mechanisms relevant to those radionuclides. It is understood that there are radio-ecological and other data and information issues which are common to specific assessments required in many countries. The mutual support within a commonly focused project is intended to make more efficient use of skills and resources, and support a transparent and traceable basis for the choices of parameter values as well as for the wider interpretation of information used in the assessments.

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

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

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

Organisation	Representative	Role of organisation	Website
Electricité de France (EDF)	Carine Damois	EDF is been the main producer of electricity in France. The Laboratoire National Hydraulique et Environnement (LNHE) department works on migration of pollutants in the ground, waste management, water quality, soil contamination, ecotoxicology, ecology, microbiology, health risk assessment, but also fluvial and maritime hydraulics, resource management, industrial flows and combustion, meteorology and air	www.edf.fr

		quality.	
Korea Atomic Energy Research Institute (KAERI)	Yong-Soo Hwang	Kaeri is developing the Korean reference concept for permanent disposal of high-level radioactive waste including spent nuclear fuel and assessing the long term post-closure safety and repository performance.	www.kaeri.re.kr
National Cooperative for the Disposal of Radioactive waste (Nagra)	Frits van Dorp	Nagra has more than 30 years experience in the development of disposal concepts for all categories of radioactive waste. Over the years, Nagra has built up extensive technical know-how and has applied this in site characterisation and performance assessment of deep geological repositories.	www.nagra.ch
Nuclear Research Institute Rez (NRI)	Ales Laciok	In the Czech Republic, NRI is the research, development and engineering organisation responsible for the development of nuclear power technologies, utilization of radionuclides and radiation in industry and medicine, and with a role to undertake fundamental research to support the long-term management and disposal of radioactive wastes.	www.nri.cz

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

General Objectives

Overall the intention is to make available the best sources of information to justify modelling assumptions. Particular emphasis is placed on key data required for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge to the environment or release from solid waste disposal facilities.

The project is driven by assessment needs identified from previous and on-going assessment projects. Where common needs are identified within different assessment projects in different countries, a common effort can be applied to finding solutions. Such solutions may readily take account of the BIOMASS Theme 1 Data Protocol, among other things.

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

The work programme has been organised in three themes:

Theme 1: Development of a Specialised Data-Base for Key Radionuclides and Process Data

Theme 2: Modelling Testing and Development Tasks

Theme 3: Site Characterisation, Experiments and Monitoring.

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

Objectives of the Model Review and Comparison for C-14 Dose Assessment Task

The objective of Task 3 within Theme 2 is to review and discuss modelling of the transport and fate of C-14 and account for the treatment of C-14 processes, pathways and accumulation in the biosphere. Particular attention is given to the appropriate choice between dynamic and equilibrium assessment models.

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

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1. INTRODUCTION, SCOPE AND OBJECTIVES

1.1 Background

Carbon, in its various forms, is the most studied of all elements. It is clearly the "building block" of life, a component of one of the dominant atmospheric gases, the principal component of material that transforms unconsolidated mineral particles into soil, and a key element in many geological systems. We have learned much of what we know about carbon from studying C-14, and we have many years of experience with C-14 as an environmental contaminant around nuclear reactors. So why do we now discuss how to model C-14?

There are perhaps two answers. The first is that in the wholesale application of IAEA Safety Series models, there was an impetus to handle C-14 in the same way as any other radionuclide, and these were traditionally treated as trace elements. For example, deposition onto leaves, weathering from leaves and translocation from leaves to other plant tissues were modelled with parameters and parameter values suitable for nuclides such as Cs-137. The actual underlying processes for C-14 are different from those for trace elements, they are: volatilization, photosynthesis and translocation of photosynthate. Doubtless the classical parameters can be modified to deal with C-14, but it is scientifically much more defensible to begin with a unique conceptual model of how C-14 will behave. The question of how best to model C-14 is within the mandate of BIOPROTA Theme 2 on Modelling and specifically of Task 3 on Model Review for C-14.

Not only do the important processes differ for C-14, but the timeframes of interest have shifted away from short time frames associated with reactor operations to longer time frames associated with geological disposal. The waste management time frame is more consistent with the time frame where natural C-14 is used for dating ancient carbon materials. The question of time frame relates to the extent of mixing when C-14 released from a source mixes with the pools of stable C in the geosphere, biosphere (terrestrial and aquatic, freshwater and marine) and atmosphere.

The use of C-14 in dating is a good example of the issue of time frame. The assumptions behind C-14 dating are that:

1. The C-14 Specific Activity (ratio of C-14 to C-12 and C-13 or stable C) of the atmosphere when the C-14 was fixed by photosynthesis in an ancient carbon sample was sufficiently uniform as to be considered constant. In other words, the global air and water media were well mixed and in isotopic equilibrium at the time the organic material or carbon sample being dating was laid down.

2. Once the C-14 was fixed in the organic materials being dated, it was completely removed from the C cycle, no more mixing occurred, and the Specific Activity (SA) remained constant (except for radioactive decay) for millennia.

From the perspective of waste management, the crux of the issue surrounding the conceptual model for C-14 becomes: when and where can mixing and isotopic equilibrium be assumed, and when and where can they not be assumed?

A complicating factor in recommending a conceptual model for C-14 may be the desire to use these same models for both contemporary operating conditions and for long-term waste management. Contemporary C-14 emissions from nuclear facilities may need to be modelled with time-dependent isotopic dilution (e.g. Chiffroy et al.

[2001]). It is outside the scope of this Task to recommend a conceptual model for contemporary conditions, however, recent work in this area was noted and referenced.

1.2   Scope and Objectives

The objective has been to review and discuss modelling of the transport and fate of C-14 and account for the treatment of C-14 processes, pathways and accumulation in the biosphere. Particular attention is given to the appropriate choice between dynamic and equilibrium assessment models.

The Task involved a review of documents detailing ecosystem models of C-14 developed or applied in the context of nuclear waste management. The appendices include related documents that have details important to the overall Task, and so were better retained in their entirety as opposed to being summarised in the main body of this report.

2. CONCEPTUAL MODEL

2.1 Underlying Principles

Radionuclide fate and transport models are generally built by following the processes and pathways that radionuclides take as they migrate through the physical space we call the biosphere and become incorporated into the bodies of biota that inhabit it. Some call this ecosystem and foodchain modelling. Many biosphere and foodchain models presently exist for tracing the movement of contaminants and radionuclides (Bergström et al., 1999; Davis et al., 1993; Klos et al., 1996; SKB, 1999; Vieno and Nordman, 1999). Their underlying principles are based on compartmentalizing the biosphere into aquatic and terrestrial components, into their physical entities such as water, air and soil and then to add in the foodchain or animal compartment for humans and animals (Figure 1). Quantities of water, air, vegetation and soil consumed are traced and along with their concentrations are mixed into the next compartment along the foodchain. A radiation dose is calculated based on quantities for ingestion (for internal dose) and timeframes for other activities such as immersion in the physical compartment as in swimming or bathing (for external dose).

Both the spatial context and the time frame are inherent in any model proposed. Thus, the dilution of any new substance, such as a radionuclide, entering any of these compartments needs to be considered in this new "mixing" pool and a new concentration for that substance calculated at each time step of the calculation or simulation. The question that we need to ask is: should a conceptual model for C-14 differ from that of any other radionuclide? The answer is that the transfers and dilution pools are similar, it is the processes and degree of mixing that differ for C-14. To further clarify this mixing, the C-14 is mixing into an isotope pool containing C-12, C-13 and C-14 (both newly introduced or already there). The mixing is not simply a volume dilution as assumed with trace elements, it is an isotopic mixing. The structure of the conceptual model may be arrived at differently for a model considering isotopic mixing, but it can consider all the same compartments as the conventional transfer or compartment model. The isotopic mixing or SA model may be simpler if, for example, there is no need to separate offal from beef or leafy vegetables from cereals. In this case, the assumption is that the offal and muscle are composed of C with a specific isotopic ratio and there is no isotopic discrimination.

Isotopic fractionation is defined as the discrimination or separation of the various isotopes, in this case of C, in a specific compartment or medium. In the discussion of isotopic mixing or the isotopic pool, attention has to be paid to how the isotopes are fractionated. It is generally assumed that there is no significant fractionation of C-14, however, it does occur. Similarly, there is fractionation of N-14 and N-15, isotopes of very similar mass [Minagawa and Wada, 1984]. Thus, any model development for C-14 must recognize this issue of isotopic fractionation in conjunction with isotopic mixing.

Natural background levels of C-14 are also important, especially for validation studies. Typically, the background levels of C-14 will far exceed those of any waste source, and so a measured SA will reflect background C-14 more than it will waste C-14.

The temporal scale for assessment modelling needs to be considered in the treatment of C-14. How does the time scale for plant photosynthesis, and the dilution with stable carbon in the plant when it takes up C-14, relate to the time scale required for the assessment of dose for C-14 travelling through the geosphere and biosphere from a nuclear disposal vault. Do the kinetics of the absorption of C-14 from plants being digested in a ruminant need to be accounted for? These and other questions related

to the temporal scale must be answered by the Task 3 Modelling Team. Since short-term predictions are not required in this context, the attention to details of the kinetics within the plant and animal and human bodies does not seem to be justified.
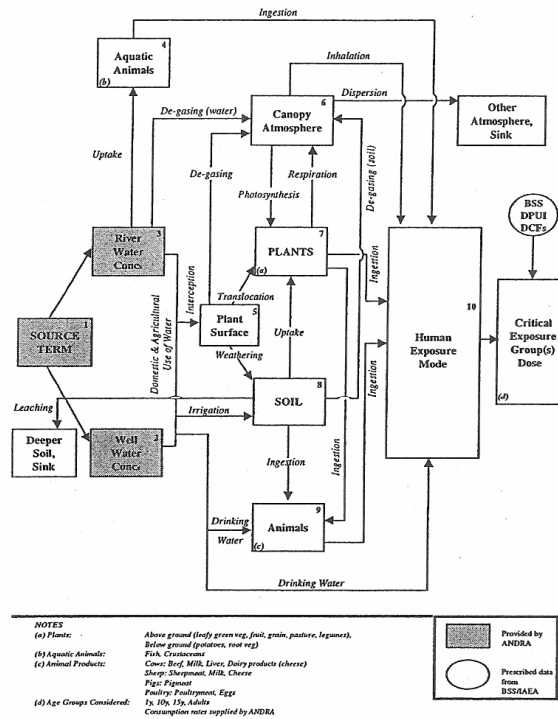
Figure 1   Conceptual model for C-14, ANDRA.

Precedence has been set for using the SA modelling approach for C-14 and for Cl-36 in nuclear fuel waste management biosphere models. We expect that since the processes that drive C cycling in the biosphere are different than for other radionuclides, the conceptual and mathematical models should differ. Is the effort to create a different model required or worthwhile? Do we have proof that we would get a different dose assessment outcome? Few comparisons have been published that allow us to answer those questions [Sheppard, 1996; Stephenson and Reid, 1996].

The SA model approach is very compatible and works well within the transfer factor models commonly employed for other radionuclides (e.g. for Cs-137, Sr-90, U-238, Tc-99, Np-237) [Davis et al., 1993]. There is probably no need for a separate model, simply a need for attention to properly construct and account for the desired mixing pools.

For C-14, Davis et al. [1993] assumed isotopic equilibrium within most local C pools.

The simplest model is to assume isotopic equilibrium in all relevant parts of the biosphere. 'Relevant' in this context is to assume isotopic equilibrium in those compartments of the biosphere that have a connection with a C-14 source and lead to dose. Perhaps the next simplest model is to assume isotopic equilibrium within a

discrete number of C pools. To consider all C pools as dynamic with continuous changes in isotope ratios could lead to a very complex model. An intermediate level of complexity, where there is always a combination of equilibrium and non-equilibrium processes being modelled, is most common and practical. One of the major present dilemmas is to decide on the most appropriate specification of this combination.

2.2   Technical Approach

The technical approach places its emphasis on determining the sizes and extent of the mixing pools for any source of C-14, whether it comes from atmospheric deposition, surface water or groundwater. If it is agreed that detailed kinetic relationships are not required for the timescale of long-term nuclear waste management, then a traditional foodchain and dose compartment model is a viable structure and the parameter values are modified to conform to the SA concept. In fact, a purpose-built C-14 SA model may look very similar to a traditional foodchain model.

Stable C concentrations are relatively constant in most organisms. Few parameter values have been derived empirically for C-14 and most rely by default on stable carbon distributions. This assumes a *de facto* SA relationship.

3. C-14 MODELLING AND METHODS LITERATURE REVIEW

The following are very brief reviews of a series of papers considered potentially relevant by the Task Group.

3.1 Carbon-14 in Low-Level Waste (EPRI)

This report offers 10 lines of text concerning biosphere behaviour of C-14, and a few pages of Appendix. They conclude that a SA model is not appropriate for groundwater release, and transfer factors are required. The basis seems to be that releases from the geosphere are point sources into the biosphere, so isotopic mixing is difficult to quantify. There is almost no scientific discussion related to C-14 in the biosphere.

3.2 Review of Reactions of Hydrogen and Methane in the Geosphere and Biosphere (Nirex)

This report described the potential attenuation of CH_4 in the geosphere and biosphere. Methane was expected to oxidise to CO_2 in the biosphere, although data for the rate of reaction were scarce. There is little else in the report to guide model development in the biosphere.

3.2.1 The Soil Plant System Model Daisy

This model included transformation and transport processes involving water, heat, C and nitrogen in the soil-plant system. The model uses a daily time step, clearly an inappropriate time frame for assessment of nuclear waste management. It does not differentiate sources of CO_2 (above ground versus below ground), which is one of the issues related to soil-to-plant transfer of C-14. Similarly, it does not differentiate within-canopy versus above-canopy sources of CO_2. This seems to be an excellent model, but largely inappropriate for waste management application.

3.3 Transfer of C-14 in the Biosphere, ANDRA, [Penfold and Watkins 1998]

This report is in three parts: a literature review, a comparison of models used by other agencies and a model proposed for ANDRA. The literature review describes a few processes in detail. Some of these are more relevant than others. There is no attempt to use literature to define and defend the level of detail needed to model C-14 for nuclear waste management. The review of models in this ANDRA report will be useful to complement the review in BIOPROTA.

The conceptual model described (Figure 2.1) is comprehensive in terms of compartments considered. It includes a plant interception-weathering-translocation process in addition to photosynthesis, and IAEA 360 is referenced. No justification was presented for inclusion of this process, and the parameter value for weathering is comparable to that used universally for non-volatile, non-photosynthetically active nuclides. This could be justified if particulate or dissolved organic carbon was envisioned as the chemical form, but this was not discussed.

The model partitions the source of C-14 to the plant between soil and canopy atmosphere, and estimates the SA in each. Similarly, it partitions the source of C to animal tissue between water, soil and plant sources. The data for this are exceptionally sparse, as almost all C in animals comes from feed. There is little

discussion of C-14 transfer to fish, which is noted in the report as a major pathway for C-14.

Overall, the model probably has a level of compartment specification that is sufficiently comprehensive, and the selection of input data can probably adapt the model to a number of conceptual models.

3.4   An Ecosystem Model of the Environmental Transport and Fate of Carbon-14 in a Bay of the Baltic Sea, Sweden (SKB), also [Kumblad et al., 2003]

This model was developed for sub-seabed disposal of C-14, and is a very comprehensive model based on observed C partitioning at the site of interest. Measured compartments were fused to achieve a manageable number of model compartments. Transfer rates among compartments were either measured or derived using simplifying assumptions. Validation was possible. A key conclusion is that all compartments had times to steady state and ecological half-times of less than 2 years. Most of the C-14 was lost from the bay by dispersion in the water column to the sea.

Specific activities varied among the compartments. The dissolved inorganic carbon (DIC) was the pool directly contaminated, and benthic plants and grazers had nearly the same SA. Most other biota had specific activities an order of magnitude lower, apparently because of dilution caused by phytoplankton uptake and transport from the model domain. The model was clearly useful to define C pools and mixing ratios.

3.5   A model for transfer of radionuclides to sewage sludge and for assessing the radiological impact of such sludge when applied to agricultural land: H-3 and C-14 [UK FSA]

This model is for C-14 in sewage sludge applied to agricultural land. The model has a series of compartments with time-dependent transfers.   In several cases, a compartment such as plants is divided in two, with a rapid-exchange and a slow-exchange (sub) compartment. The results showed that steady-state concentrations were reached in about 4 years, with the exception of animal tissue. Clearly, the times to steady state are very short compared to the time frames for waste disposal. None the less, the partitioning among compartments from such a model would be of use.

3.6   Modelling the Transfer of Tritium and C-14 in Terrestrial and Aquatic Ecosystems Discharged to Water Courses [EDF, 2001]

This model, presented by Ciffroy et al. [2001] and reviewed by Sheppard and Stephenson [2002], is a dynamic model intended to predict concentrations in the food chain for C-14 in river water that varies in concentration on a time scale of month to month. It is a full SA model, and for each compartment it computes the change in SA as a function of time. The critical parameter is a turnover rate, and this encompasses processes such as biochemical turnover within an organism, growth of an organism or the population, and turnover as a result of death and decomposition. The data for these turnover rates are not plentiful.   The model also accounts for isotopic discrimination, although the effect is rather small. The turnover rates in some biota are slow enough that changes in substrate C-14 concentrations on the order of months are effectively dampened.   However, for changes in substrate C-14 concentrations on the order of decades or centuries as might be expected in a waste management scenario, the rate of turnover is fast enough to be considered at

equilibrium. Validation of this model is underway [F. Siclet, personal communication]. An interesting aspect of the model is the diffusive loss of C-14 from the water to the atmosphere: the concentration gradient for stable C is often from air to water, whereas the concentration gradient for C-14 is from water to air. The net result depends to a large extent on the role of algal photosynthesis in the fixation of C-14 before it can escape the water by diffusion.

3.7 Assessment of the impact of IRUS on Duke Swamp [Sheppard and Reid, 1996]

A proposed Intrusion Resistant Underground Structure (IRUS) for the disposal of low-level radioactive waste, containing C-14 among other radionuclides, was assessed. Since it was proposed to be constructed in a sandy ridge upslope of a swamp, an assessment of the transport and fate of the proposed radionuclide inventory through the subsurface aquifer to the wetland and beyond was undertaken. Concentrations in swamp water, peat and vegetation were predicted as inputs to estimates of both internal and external doses to humans, using a traditional transfer factor model structure. Special attention was given to the plant concentration factor (Bv) to ensure its value accounted for stomatal uptake from a dispersing atmospheric plume, as well as through the groundwater-soil-plant pathway. Carbon-14 gave the second highest predicted dose (H-3 gave the highest) and this dose was attributed to ingestion of vegetation (berries from the swamp or produce grown in a garden with soil ameliorated with peat from the swamp).

3.8 Fate and distribution in sediments of carbon-14 added to Canadian Shield lakes of differing trophic state [Stephenson et al., 1995]

Carbon-14 was added to several lakes in the Experimental Lakes Area of Canada in 1976 and 1978 to study the primary production and gas exchange with the atmosphere. This resource was then included in studies of the long-term fate of C-14 for the nuclear fuel waste management program. The authors showed that 29% of the C-14 still remained in the lake; however, some 9% of this was mineralized. Sediment focusing caused the redistribution of C-14 in this lake for 2 to 7 years following the addition. After 11 years, Lake 226N retained about 18% of the original C-14, with 13% of that remaining in erosional areas of sediment and 87% in transitional or depositional areas. Similar distributions were found in the mesotrophic and oligotrophic lakes (Lake 224 and 226N), but the retained total was lower (about 7%). This study highlights the effect of chemical speciation on C-14 behaviour: as inorganic C-14 it was rapidly lost, but any that was captured by photosynthesis in the lake persisted in the sediment.

3.9 Carbon-14 activity in the water, sediments and biota of Lakes 226 North, 226 South and 224, Experimental Lakes Area, 1989 to 1994 [Stephenson et al., 1994]

This work reported further data for Lakes 224, 226N and 226S at the Experimental Lakes Area; data for 10 to 15 years after the C-14 introduction. Data provided included particulate carbon, dissolved inorganic carbon (TIC), dissolved organic carbon (TOC) and biotic C-14 (whitefish). The data suggest that SA is constant with depth in the surface sediment, but in the buried-depositional zone sediments there is a peak in the C-14 activity and the SA between 4 and 5 cm below the sediment-water interface, an important piece of information for construction of a system model. As illustration of the relationship between a SA and a trace element approach, comparison was made between C-14 and Co-60. Carbon-14 peaks were tighter than

peaks of Co-60 used as a metal tracer and spiked at the same time. This suggested that C-14 incorporated into organic particles had less post-depositional mobility than Co-60. Both TIC and TOC showed C-14 activity increasing with depth in the water column, indicating that C-14 enriched sediments are still decomposing, more than a decade after deposition, and releasing C-14 to the surface waters. The study also showed that loss of -C-14 through gaseous exchange with the atmosphere caused the primary production food web to have a lower SA of C-14 than the detrital food web. A time-series collection of fish indicated rapid turnover of C in whitefish (~1/y), but much slower turnover in lake trout muscle tissues.

3.10 Validation test for carbon-14 migration and accumulation in a Canadian Shield lake [BIOMOVS II, 1996]

This validation test presented by BIOMOVSII used the data in the earlier report by Stephenson et al. [1994]. The data were said to be useful for the investigation of both short-term processes relevant to the evaluation of accidental releases as well as longer-term processes relevant to routine releases and to solid waste disposal. Four international models participated in this validation scenario and the predicted endpoints were C-14 concentrations in water, sediment and whitefish over a thirteen year period. All models gave reasonable estimates of the water and sediment endpoints. It was noted that the transfer rate to sediment and the gaseous evasion rate from the water to the atmosphere are important in influencing the predictions. Large uncertainties in the fish concentrations, together with the tendency for all the models to underestimate them, emphasized the need to have a better understanding of the processes that influence contaminant concentrations in fish. The dynamic models predicted the fish concentrations better than did those models using equilibrium concentration ratios. All the models failed to accurately predict the C-14 values in fish; they tended to overestimate the loss of C-14 from fish.

3.11 Relative importance of atmospheric and root uptake pathways for $C(14)O_2$ transfer from contaminated soil to plants [Amiro et al., 1991]

This paper estimated and compared the relative C-14 fluxes from the atmosphere above an area source to vegetation and to plants by direct root uptake. The contribution from the atmosphere depends on the rate of volatilization from the soil, the size of the source area and the meteorological dispersion conditions. The calculations were performed using an atmospheric dispersion relationship and showed that the ratio of atmospheric to root uptake pathways depended on plant height and source area, exceeding unity when the fetch was greater than a few meters for bean plants, but much greater for maize. A very large fetch, greater than 1000 m, is required before a uniform SA throughout the local biosphere can be assumed for a soil source. This quantitative analysis describes the limitations for calculations and experiments used to define simplified transfer coefficients for C-14 from soil to vegetation and shows that the relative importance of the pathways depends on the geometry of the source. It also indicates how important it is to model (or otherwise deal with) the spatial extent of contamination resulting from a waste disposal facility.

3.12 Carbon-14 in the biosphere: modelling and supporting research for the Canadian Nuclear Fuel Waste Management Program [Sheppard et al., 1994]

The key message of this report on modelling C-14 in the biosphere is the use of a partial isotopic exchange approach, with the remainder of the model relying on compartmental transfers. The paper suggests that the soil-atmosphere to plant

pathway dominates and the gaseous loss of C-14 from soils and lakes is significant. The authors also note that recalcitrant forms of C-14 may persist in soils and sediments for long periods. The relative impact of these forms is low because their bioavailability is also very low. They also state that full isotopic exchange will not be achieved in small areas or in short time intervals. Degassing into the atmosphere and flushing of water bodies are the major mechanisms of loss from the local biosphere. One application of the SA concept employed in this program is to set an upper limit on C-14 dose corresponding to the highest possible SA in the biosphere (well water in this case).

3.13 Towards a model for the dynamic transfer of tritium and carbon in mammals [Galeriu et al., 2003]

Available data from experiments with rats and sheep have been analysed to test the hypothesis that both H-3 and C-14 transfer in mammals can be accounted for by an understanding of metabolism. A multi-component retention function has been developed for various organs. Allometric equations have been derived for muscle half-times. The function could be used in the development of a human biokinetic model. Several points were made. For the whole body or a major organ, at least two components of loss are required; one with a short and another with a long half-time. The authors considered that mass-dependent relationships for C-14 loss rate can be defined across species.

3.14 Technical description of a model of the carbon cycle in Canadian Shield lakes [Stephenson and Reid, 1996]

A model of the carbon cycle in Canadian Shield lakes was developed and tested using field observations on the fate of experimentally added C-14 in three lake basins (see other Stephenson papers). The model was calibrated over a 16 year period using L226 north basin data and validated by applying it to the south basin of the same lake (L226) and another (L224). The validation and calibration success suggests this model can be used to predict the fate of C-14 and pathways of C transport in lakes. This model is based on the dynamics of C-14 in the dissolved inorganic carbon (DIC), dissolved organic carbon (DOC) and particulate organic carbon (POC) pools of both the epilimnion and the hypolimnion and these processes are followed through annual cycles of ice cover and twice-yearly mixing of the stratified water body. Transfers of contaminants are described as first-order processes and flushing of contaminants to a stream outflow is driven by hydrological data. This model is not a full SA model because the amounts of stable C in the mixing pools are not used for every calculation: however, it is somewhat equivalent to a SA model because the transfer parameters involving C were based on data for stable C and were not unique to C-14.

A comparison was made between this model and that of the steady-state BIOTRAC surface water model [Bird et al., 1993]. The surface water model permits only one aqueous and one sediment compartment and exchange is limited to the unidirectional net flux of water to sediment. Losses to outflows, radioactive decay and gaseous evasion to the atmosphere are also modelled in the surface water model, but these sinks for C-14 are not treated as compartments. In this dynamic model, there are multiple compartments that process C at different rates, reflecting known physical, chemical and biological processes. This dynamic model also includes seasonal effects by simulating the annual mixing cycle in the water column, sediment resuspension and seasonally variable transport rates between compartments. The different predictive capabilities of the two models are a direct result of the structural sophistication and simulation timescale. For long-term net exchange between water and sediment, assuming a continuing source-term flux to the water, the steady-state

surface water model of Bird et al. [1993] and the dynamic C-14 model converge to very similar values. More discussion of the features and results of the dynamic model are given in the Stephenson document.

3.15 Specific Activity models in relation to BIOTRAC biosphere model [Sheppard, 1996]

This document, appended in its entirety as Appendix A, starts the discussion of SA models by saying that SA models apply to radionuclides where the stable element is abundant and considers the behaviour of the stable plus the radioactive isotopes of the element. It is intended as a primer for those needing more explanation of the concept of SA. Some key points are excerpted here.

It is noted that because C-14 is abundant in the environment and is biologically essential, that a contaminant fate model employing transfer factors, as used in determining the fate of trace elements, may not be appropriate. For example, the concentration of C in biota, as mass per unit dry weight of biota, has narrowly defined limits. A transfer factor model for C-14 would imply continued accumulation of C-14 regardless of this limit for C accumulation, whereas a SA model would specify the C-14/C-12 ratio for the defined amount of C-12 in the biota. The question the report addresses is whether isotopic dilution is adequately modelled in the transfer model BIOTRAC. For dose estimation, the concept of a SA model includes the compartment or mixing pool accessed by the critical group, but this receptor may access several mixing pools and not be in isotopic equilibrium with any one. This is when the SA model becomes complex.

One of the simplifying assumptions often made in a SA model is that since plants obtain all their C from the atmosphere, for C-14 released to the atmosphere, the SA of C in the plants is equal to that in the atmosphere and the discrimination between isotopes is nil. This is because the atomic weight of C-12 and C-14 differ by only 17% whereas more discrimination could be expected for H-3 and H-2 where the atomic weights differ by 50%. In this document, Sheppard discusses how SA model components for C-14, Cl-36 and I-129 are already part of BIOTRAC. For C-14, two uses are made of the SA concept, the first is the use of the geosphere SA to place an upper bound on the dose estimate and the second is related to the fact that several parameter values for C-14 are based on ratios for the stable element. If the C-14 transfer parameters are based on stable C the result is a SA model. This was the case for the soil-to-plant and water-to-fish transfer values in BIOTRAC.

Sheppard [1996] explored the time-dependent approach to isotopic equilibrium in soils and concluded that a more detailed SA model for C-14 in soil, taking into account the more recalcitrant forms of C, will not significantly change the dose estimation. This was also shown by Amiro et al. [1995] in his population dose estimate using a simple SA model.

3.16 Specific-Activity relationships for calculating doses from irrigation with contaminated groundwater: The case of I-129, C-14 and Cl-36 [Amiro, 1996]

The author answers the question "What is a reasonable SA to use for calculating doses to humans from an irrigation scenario, since some of the radionuclide and stable element will be lost to the atmosphere or leached from the soil? Further, the C-14 only originates from the contaminated irrigation water, whereas the stable isotope concentration is the sum of the input from the irrigation water source and the resident inventory in the soil irrespective of irrigation.

Analysis of the final SA of C-14 in the soil, as a result of irrigating with contaminated water, and the time at which this final SA is achieved, depends on the loss rate constant, k_j, or leaching rate. If realistic leaching rate constants are used, then the soil comes to a constant SA quickly, in a few years. If the loss rate is much smaller, such as 10^{-3}/y, then it will take over 1000 years before a constant SA, the maximum SA based on the groundwater, is reached in the soil for C-14. This analysis also shows that the SA in soil from irrigation, using realistic leaching rates, is three orders of magnitude below the maximum value based on the SA of the irrigation water.

3.17 Stepwise enrichment of N-15 along food chains: Further evidence and the relation between δN-15 and animal age [Minagawa and Wada, 1984.

The isotopic composition of nitrogen was measured in marine and freshwater animals from the East China Sea, the Bering Sea, Lake Asinoko and the Usujiri intertidal zone. All consumers, zooplankton, fish and birds exhibited stepwise enrichment of N-15 with increasing trophic level. A similar N-15 enrichment occurs in all molluscs, crustaceans, insects, amphibia, fish, bird and mammal species, regardless of the difference in the form of excreted nitrogen, and in laboratory cultured fish, brine shrimp and mice.

These data show that N-15, comparable in mass to C-14, is fractionated during the feeding process, and the authors mention that the fractionation factor is "more than those of C and H". The authors summarize with: "the enrichment of N-15 is widespread among most animals collected from different ecosystems and isotopic enrichment occurs independently of habitat, form of N excreted and growth rate. The N isotopic composition of these animals is influenced however by the ultimate source of the fixed N".

4. CONCLUSIONS AND RECOMMENDATIONS

4.1 Issues

◆ The crux of the issue surrounding the conceptual model for C-14, from the perspective of waste management, becomes: When and where can mixing and isotopic equilibrium be assumed, and when and where can they not be assumed? This must be resolved with a full understanding of the system to be modelled and carefully specified in the conceptual model and scenario.

◆ Definition of the temporal scale for the assessment, and in conjunction with the timescale, decision as to whether kinetic models are useful and/or necessary.

◆ Understanding of the implications of fractionation of C-13 and C-14 with respect to C-12 in the biosphere physical compartments and at the various trophic levels of both the aquatic and terrestrial systems.

4.2 Knowledge Gaps

◆ More data are needed for gaseous exchange from surface water and soils to the atmosphere and how this affects the SA in the water and soil compartments.

◆ SA and concentration values in animal tissues with time are required, and investigation is needed as to what is the turnover rate that determines the change in SA.

◆ More knowledge of stable isotope concentrations in well and groundwater would enhance the prediction capabilities of simple SA models.

4.3 Technical Approach

◆ Major components (lake, soil, atmosphere, plant) of a biosphere model developed as a SA model exist to be knitted together.

◆ A traditional trace element food chain model can be converted to a full SA model, without the need for a newly developed separate SA model.

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1. APPENDIX A. SPECIFIC ACTIVITY MODELS IN RELATION TO BIOTRAC BIOSPHERE MODEL.

See separate pdf documents on the Business Collaborator website.

2. APPENDIX B. THE IMPLICATIONS OF RELEASES OF C-14 IN GASEOUS FORM FROM A RADIOACTIVE WASTE REPOSITORY

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1. INTRODUCTION

Gaseous forms of C-14 can be evolved as a result of waste degradation in a deep geological repository for radioactive wastes. In the context of a UK repository for low and intermediate-level radioactive wastes, the principal forms of gaseous C-14 likely to be evolved are labelled carbon dioxide and methane, though ethylene and acetylene may also be produced. A review of issues arising from gas releases to the accessible environment in the transport, emplacement, operational and post-closure activities and phases of a deep geological facility for the disposal of low- and intermediate-level radioactive wastes has recently been undertaken for United Kingdom Nirex Limited [Thorne and MacKenzie, 2004]. That review includes specific consideration of issues relating to C-14. It was considered that it would be useful to describe aspects of that report relevant to C-14, as these are likely to be of general interest to participants in BIOPROTA. In view of the remit of BIOPROTA, only aspects relating to post-closure behaviour are discussed below.

2. IMPACT OF GASEOUS C-14 IN THE NIREX GENERIC PERFORMANCE ASSESSMENT

2.1 Gas Production

In a generic post-closure performance assessment (GPA) that has recently been undertaken by Nirex [Nirex, 2003], the following assumptions were made concerning gas production.

The sources of non-radioactive gas are:

a) Hydrogen production from anaerobic corrosion of mild and stainless steel, Zircaloy and uranium in the wastes, of mild and stainless steels making up containers, and of stainless steel in stillages;

b) Hydrogen production from corrosion of the active metals Magnox and aluminium;

c) Gases (carbon dioxide, methane, hydrogen, nitrogen and hydrogen sulphide) produced by the microbial degradation of organic materials or from reactions of sulphate and nitrate ions; and

d) Hydrogen production by radiolysis of water.

In addition, the production of the following radioactive gases was considered:

a) Tritiated hydrogen from the corrosion processes, either by release of tritium in the metals or by incorporation of tritium present in water in the wastes into the gaseous hydrogen produced;

b) C-14-substituted methane and carbon dioxide by incorporation of C-14 in the organic wastes into the microbiologically generated gases;

c) Rn-222 from the decay of Ra-226.

It was considered that the result of reactions between groundwater containing dissolved carbon dioxide and cement material in the waste container and the backfill would be the precipitation of calcium carbonate; any carbon dioxide was therefore expected to become effectively immobilised. However, it should be noted that there is the potential for carbon dioxide to migrate along cracks in cement where the surfaces have already been carbonated, so some issues still remain as to whether carbon dioxide and hence the C-14 incorporated in carbon dioxide would be completely trapped in the engineered system.

There are other potential sources of C-14 in gas that could usefully be investigated further in relation to individual waste streams. To date, the other potential sources of C-14 in gas that have been identified as worthy of further study include:

- C-14 in the form of metal carbides, which may be converted into acetylene or methane gas;

- C-14 -bearing gas originating from graphite in the waste inventory.

The C-14 in graphite could be efficiently trapped within the very stable structure of the graphite, or it could migrate along grain boundaries and be released.

Gas generation calculations were undertaken using the code GAMMON [Agg et al., 1997]. In addition, supplementary calculations were undertaken for processes not included in GAMMON. These comprised:

◆ Gas generation as a result of radiolysis of conditioned wastes;

◆ Gas generation by metallic uranium corrosion; and

◆ Rn-222 generation and release.

Within GAMMON, microbial reactions between carbon dioxide and hydrogen to form methane can be simulated. In the reference calculation, this process was deactivated to simulate the effects of carbonation, which would remove carbon dioxide from the system. The preferential reaction of carbon dioxide with cements preventing the formation of methane is significant, because without it C-14 -labelled methane formed from C-14 -labelled carbon dioxide would not be trapped by the carbonation reaction. A variant study was undertaken in which the reaction between carbon dioxide and hydrogen was permitted.

In addition, two variant studies were undertaken to investigate the effects of repository operating and closure histories on the production of bulk and active gases. Variant 1 considered the effect of early vault backfill followed by a period of care and maintenance before final closure. Variant 2 considered the case of early backfill and immediate closure. Variant inventories were also considered.

2.2 Gas Migration

There are two main mechanisms by which gas could migrate through the geosphere: either in solution in the groundwater or as a free gas phase [Nirex, 1994]. Studies of typical situations have shown that neither the diffusive flux nor the advective flux of dissolved gas is likely to be sufficient to carry away, from a repository, all of the gas that would be generated within it [Rodwell and Nash, 1991]. As a consequence, a free gas phase is expected to exist and gas will migrate through the geosphere in this form. As a free gas phase, gas may move as bubbles or as a continuous gas stream, or as a mixture of both. In the case of bubbles, the flow is driven by buoyancy forces and would therefore be predominantly in the vertical direction. For gas flow in a continuous stream, both buoyancy and the pressure of the gas will drive the gas migration. Pressure-driven flow could also produce lateral gas migration.

Gas would move either through the rock pores or preferentially through fractures that could exist in certain parts of the geosphere. The relative magnitudes of the contributions of the different mechanisms (*e.g.* as a continuous gas stream or as bubbles) will depend on the gas generation rate and the properties of the rock through which the gas passes.

As the gas in a free gas phase moves away from a repository, it may dissolve in groundwater. The extent to which this might occur would depend on the magnitude of the groundwater flow compared to the gas generation rate and the extent to which the gas migrating to the surface comes into contact with flowing groundwater. Where groundwater saturated with dissolved gas moved towards the surface, the associated reduction in hydrostatic pressure might result in gas coming out of solution and forming a free gas phase.

Thus the exchange of gas between the liquid and gas phases could form an important part of the gas pathway through the geosphere.

In the generic post-closure performance assessment, simple one-dimensional gas migration calculations were undertaken using the software TOUGH2 [Pruess, 1987; 1991]. The gas migration model was used to simulate the post-closure period, so the initial conditions related to the state of the repository at closure. A continuum model of two-phase flow in a porous medium was used to compute the resaturation and subsequent build-up of gas pressure in the repository, and to provide rough estimates of the timescales for the eventual escape and migration of gas from the repository towards the surface. It was assumed that there was no flow through the sides of the model, and that the base was a constant pressure boundary. In the reference case assessment of the gas pathway, a hard, fractured host rock was assumed. A sensitivity study was also undertaken, in which the host rock was assumed to be a very low permeability argillaceous (clay) stratum.

Two hydrogeological units were assumed in the reference case assessment of the gas pathway: a hard, fractured host rock overlain by a fairly low permeability rock.

2.3 Impact Modelling

Analytical models were used to evaluate the following potential impacts to representative members of the potential exposure group:

◆ The radiological impact from tritium in the biosphere, including inhalation, uptake through skin and ingestion;

◆ The radiological impact from C-14 in the biosphere, including inhalation and ingestion;

◆ The radiological impact from Rn-222 by inhalation.

Analytical models were also used to assess the potential for flammable gas mixtures (*e.g.* hydrogen/oxygen) to accumulate in confined spaces in surface buildings.

Breakthrough calculations undertaken with TOUGH2 for the reference case suggested that the timescale for the gas pathway to form was about 6,000 years. Although a delay of 6,000 years would have a significant effect on the inventory of C-14 available for release, it would only reduce it by about a factor of two. In the reference case model, essentially all the C-14 incorporated in carbon dioxide was considered to react with the grout inside the waste containers. Therefore, consideration had to be given only to releases of C-14-labelled methane. It was assumed that all such methane generated in the repository was discharged immediately into the biosphere. The peak generation rate was estimated at $\sim 1 \; 10^{-4}$ TBq/y, occurring at about one hundred years after closure. The main radiological impact from release of the gas was found to arise from the ingestion route, due to plant uptake of the radionuclide from the atmosphere after its transformation from methane to carbon dioxide in the soil zone (see below).

With the peak generation rate of C-14 of $\sim 1 \; 10^{-4}$ TBq/y and a release area of 279,600 m^2 (the plan area of the relevant vaults, including pillars of host rock separating adjacent vaults) the effective dose rate to the most exposed individual was calculated as $1.49 \; 10^{-7}$ Sv/y, corresponding to an annual risk of $8.96 \; 10^{-9}$. However, a smaller area of discharge could be appropriate, if the gas discharge was restricted to a few fractures. For a release area of 10,000 m^2, the peak effective dose rate was calculated as $7.90 \; 10^{-7}$ Sv/y, corresponding to an annual risk of $4.74 \; 10^{-8}$. Of course, the release area could also be larger than the repository footprint, but this would not be considered to provide a reasonably cautious basis for assessment.

Risks from inhalation in buildings were found to be significantly smaller than those from discharge to soil.

In the variant calculation in which the reaction of carbon dioxide with hydrogen to produce methane was permitted, the rate of production of C-14-labelled methane increased from ~ 1×10^{-4} TBq/y to ~1.0 TBq/y in the first 100 years after closure. For a limited release area of 10,000 m^2 this results in an increase in the peak annual risk from 4.74×10^{-8} to ~5×10^{-4}. However, dispersion and radioactive decay could reduce this risk over the timescale of 6,000 years estimated as required for the gas pathway to develop.

The biosphere calculations relating to releases of radioactive gases are not described in detail in the generic post-closure performance assessment. However, the methodology used has been described elsewhere [Thorne, 2003a].

The methodology and parameter values were taken, as far as possible, from Volume 4 of Nirex 97 [Nirex, 1997]. However, some modifications were made.

For indoor exposure, the relevant methodology is set out in Section 5.1 of Volume 4 of Nirex 97. It is assumed that a building of plan area A_B is present within the area of the release, A. The fraction of the release entering the building, f, is given by:

$$f = \eta A_B / A \qquad \text{for} \qquad A_B < A$$

$$f = \eta \qquad \text{for} \qquad A_B \geq A$$

subject to the constraint that f lies on the range [0, 1]. The quantity η is an enhancement or depletion factor, to allow for the preferential ingress of contaminated air into, or its exclusion from, the building.

The concentration of any radionuclide in the air of the building is given by:

$$C = fR / V(\lambda_V + \lambda_D + \lambda_R)$$

where C (Bq/m^3) is the concentration of the radionuclide of interest in the air of the building;

R (Bq/y) is the release rate of the radionuclide to area A;

V (m^3) is the volume of the building;

λ_V (1/y) is the air exchange rate of the building;

λ_D (1/y) is the depletion rate of the radionuclide due to deposition onto surfaces within the building;

λ_R (1/y) is the radioactive decay constant of the radionuclide.

Values of λ_R are computed from the radioactive half life, T_R (y) using:

$$\lambda_R = \ln(2)/T_R.$$

Finally, values of annual effective dose, H (Sv), are calculated using:

$$H = CO_{indoor}H_B$$

where O_{indoor} is the fractional indoor occupancy;

H_B (Sv/y per Bq/m^3) is the dose rate from exposure to the radionuclide of interest at unit concentration in air.

Reference parameter values and their bases are summarised in Table 1.

Thus, the only remaining quantities required are the values of effective dose rate for unit concentration in air. These are listed for C-14 in Table 2, together with their derivations.

Table 1: Parameter Values used in Indoor Gas Exposure Calculations in the Generic Post-closure Performance Assessment undertaken by Nirex

Quantity	Value	Comment
A	1E+06	From Nirex 97.
A_B	68	In Nirex 97, it was argued that the key parameter is V/A_B. This was taken as 2.5 m, the height of a typical room. As V was taken as 170 m^3, this implies that A_B was 68 m^2.
η	0.5	From Nirex 97, based on a limited effectiveness of floors in excluding radioactive gases. However, it was noted that values of η could vary from <0.01 for buildings with intact floor slabs to ~ 1.0 for wooden-floored buildings.
R	1.0	From Nirex 97, normalised release rate.
V	170	From Nirex 97.
λ_V	8766	Value of 1/h, consistent with the recommendations for human intrusion and general data on air exchange rates in buildings.
λ_D	0	As discussed in Nirex 97, this is negligible compared with λ_V and can be neglected.
O_{indoor}	0.95	From Nirex 97.
T_R(H-3)	12.3	From guidance issued by Nirex on values to be used for the GPA, but also consistent with Chu *et al.* [1999].
T_R(C-14)	5730	

Table 2: Effective Dose Rates for Unit Concentrations of C-14-labelled Gases in Air

Gas	Effective Dose Rate for Unit Concentration in Air (Sv/y per Bq/m^3)	Derivation
C(14)H$_4$	9.00E-10	As in Nirex 97, as no updated information is available.
C(14)O$_2$	4.00E-08	Based on a committed effective dose per unit intake value of 6.2 10^{-12} Sv/Bq from Table A.3 of ICRP Publication 72 [ICRP, 1996] and an inhalation rate of 6500 m^3/y. Rounded to two significant figures.

As discussed in Nirex 97, both C(14)H$_4$ and C(14)O$_2$ entering the soil zone can be treated as if they were C(14)O$_2$, as a consequence of the quantitative microbiological metabolism of C(14)H$_4$. The implications of release of C(14)O$_2$ to the soil zone were studied in detail using the RIMERS model [Nirex, 1994] and equation 5.19 of Volume 4

of Nirex 97 is a regression relationship based on an analysis of the results of sensitivity studies undertaken with that model. This regression relationship is:

$$C_P = 1.13(M_{10}/8.84\ 10^{-4})^{-1}(\lambda_{1,9}/0.70)^{-0.4}(\lambda_{10}^L/1000)^{-1}$$

where C_P (Bq/kg(C)) is the concentration of C-14 in plants for a release rate of 1 Bq/m^2/y to the root zone;

M_{10} (kg/m^2) is the mass of stable carbon per unit area in the above-canopy atmosphere;

$\lambda_{1,9}$ (1/y) is the rate coefficient for transfer of carbon from plants to the below-canopy atmosphere;

λ_{10}^L (1/y) is the loss rate from the above-canopy atmosphere.

The normalisation values for M_{10} and $\lambda_{1,9}$ in the equation for C_P are taken to be typical values of the associated parameters. The value of λ_{10}^L is calculated explicitly using:

$$\lambda_{10}^L = n_S kv/A^{0.5}$$

where n_S is the number of seconds in a year (3.156 10^7);

k is a geometric factor relating to the contaminated area and takes a value of 0.752 for a circular area [Thorne, 2003a];

v (m/s) is the mean wind speed (see below).

As pointed out in Nirex 97, it is the reciprocal of the windspeed that determines C_P. Therefore, v is properly computed using:

$$v = 1/\Sigma(f_i v_i)$$

where f_i is the fraction of the time that Pasquill Stability Category i occurs;
v_i (m/s) is the mean wind speed in Pasquill Stability Category i.

Values of f_i and v_i are given in Nirex 97.

Thus, the concentration of C-14 in plants, C_{plant} (Bq/kg(C)) is given by:

$$C_{plant} = RC_P/A.$$

Using a specific activity argument the concentration of C-14 in animal tissues is expected to be similar. Therefore, the annual committed effective dose to the representative member of a potentially exposed group can be calculated using:

$$H = 365.25\,C_{plant} I_{carbon} H_{ing}$$

where H (Sv) is the annual committed effective dose;

I_{carbon} (kg/d) is the ingestion rate of carbon in diet (taken as 0.3 in Nirex 97);

H_{ing} (Sv/Bq) is the committed effective dose per unit intake for dietary C-14 (taken as 5.8 10^{-10} from ICRP Publication 72 [ICRP, 1996]).

2.4   Overall Conclusions

Overall conclusions from the post-closure generic performance assessment relating to gaseous releases of C-14 were the following.

◆ For the reference case scenario, the rate of release of C-14 -bearing gas is too low to cause a radiological hazard from release at the surface that would approach the regulatory target for such releases. Risk would be reduced further if C-14 - bearing gases were released over a larger area than assumed in this study; however, release over a smaller area would result in an increased risk.

◆ Significant C-14-bearing gas release is almost entirely confined to the variant scenario in which carbon dioxide and hydrogen are allowed to react in the vaults. For this no-carbonation variant scenario, a short period during which there is a high rate of C-14-bearing gas generation is calculated. Release to the surface at this generation rate would cause doses and risks to potentially exposed individuals that would breach the regulatory target. Were instantaneous breakthrough to occur, specific measures might be needed, on a waste stream by waste stream basis, to eliminate biodegradable forms of C-14 or ensure their degradation was inhibited by some means. Such an approach would identify if specific measures were needed to ensure the acceptability of a waste stream for disposal.

◆ The differences between the results for the scenarios involving different repository operational and closure histories are rather small and predictable.

◆ The differences between results obtained with the different inventories can be explained straightforwardly in terms of the different quantities of gas-generating materials in the inventories. However, the variant volume inventory includes a significant increase in the amount of graphite being packaged and coming forward for disposal. This graphite contains a significant inventory of C-14, which could have a major impact on the gas pathway were it to be released in gaseous form. The chemical form and release characteristics of C-14 in graphite under saturated alkaline conditions is an area where further work is required, in order to provide data and justification for the approach to be pursued in a future performance assessment.

3. THE C-14 MODEL DEVELOPED BY THE UK FOOD STANDARDS AGENCY

The UK Food Standards Agency (FSA) has recently developed models for the uptake of H-3 and C-14 by soils and plants after the addition of these radionuclides to the soil zone in agricultural fertilizer products based on sewage sludge [Thorne et al., 2003a]. These models are directly applicable to post-closure releases of these radionuclides in gaseous form to the soil zone and can also be adapted to assessments of the radiological impacts of releases to atmosphere. A brief account of the C-14 model is provided below and a comparison with the approach used by Nirex is provided in Section 5.

3.1 Model Structure

The overall structure of the model adopted to represent addition of C-14 to agricultural land is shown in Figure 1.
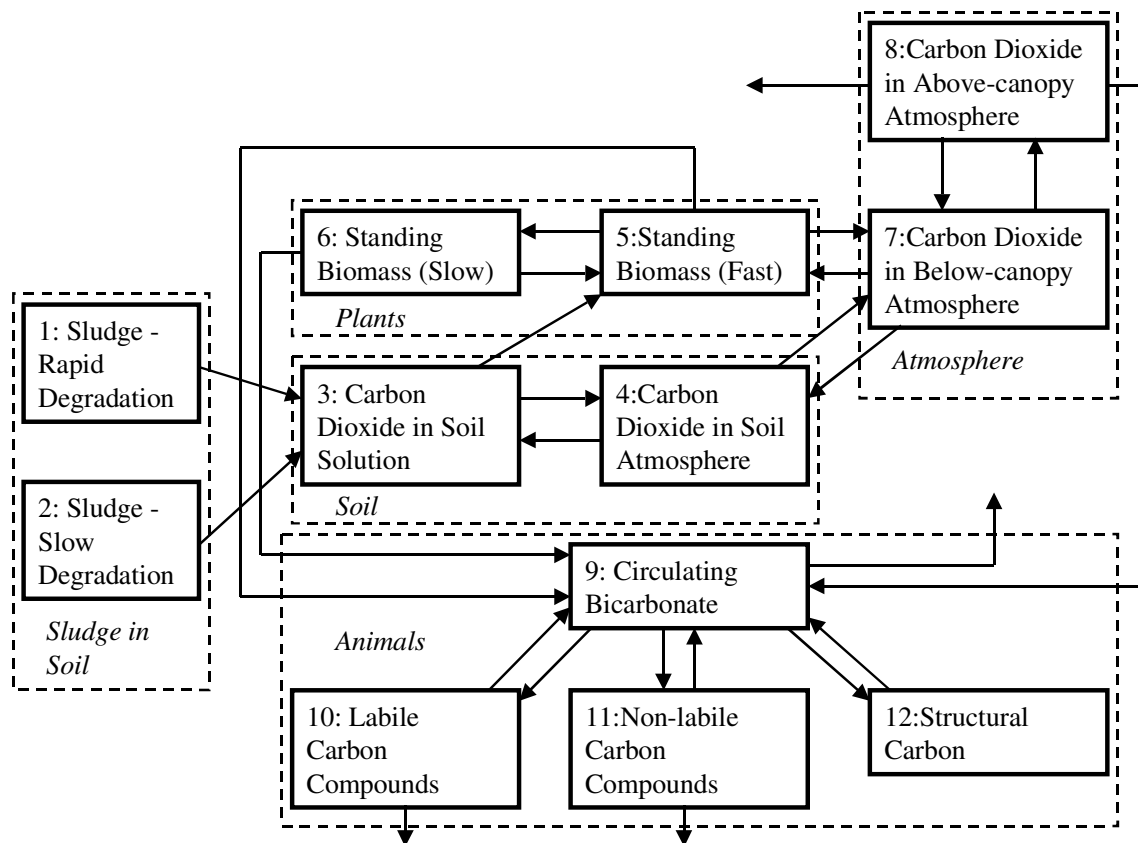
Figure 1: Structure of the FSA C-14 Model

Using obvious notation, the basic equations of the model are as set out below.

$dq_1/dt = I_1 - (k_{13} + k_R)q_1$

$dq_2/dt = I_2 - (k_{23} + k_R)q_2$

$dq_3/dt = k_{13}q_1 + k_{23}q_2 + k_{43}q_4 - (k_{34} + k_{35} + k_R)q_3$

$dq_4/dt = k_{34}q_3 + k_{74}q_7 - (k_{43} + k_{47} + k_R)q_4$

$dq_5/dt = k_{35}q_3 + k_{65}q_6 + k_{75}q_7 - (k_{56} + k_{57} + k_{59} + k_R)q_5$

$dq_6/dt = k_{56}q_5 - (k_{65} + k_{69} + k_R)q_6$

$dq_7/dt = k_{47}q_4 + k_{57}q_5 + k_{87}q_8 - (k_{74} + k_{75} + k_{78} + k_R)q_7$

$dq_8/dt = k_{78}q_7 - (k_{87} + k_{89} + k_{8L} + k_R)q_8$

$dq_9/dt = k_{59}q_5 + k_{69}q_6 + k_{89}q_8 + k_{109}q_{10} + k_{119}q_{11} + k_{129}q_{12} - (k_{910} + k_{911} + k_{912} + k_{9L} + k_R)q_9$

$dq_{10}/dt = k_{910}q_9 - (k_{109} + k_{10L} + k_R)q_{10}$

$dq_{11}/dt = k_{911}q_9 - (k_{119} + k_{11L} + k_R)q_{11}$

$dq_{12}/dt = k_{912}q_9 - (k_{129} + k_R)q_{12}$

In this model, allowance is made for two components of degradation in soil. In these equations, I_1 and I_2 are the rates of input of activity to the fast and slow degradation compartments, respectively. They are defined using:

$I_1 = M_{add} \, f_{fast} C_{product}$

$I_2 = M_{add}(1 - f_{fast}) C_{product}$

where M_{add} (kg/m^2 or kg/m^2/y; depending whether spike or continuous input is considered) is the amount or rate of addition of the product to the system per unit area;

f_{fast} is the fraction of the added activity associated with the rapid degradation component; and $C_{product}$ may either be the concentration at some particular time, or a weighted average value, e.g. if stocks of the product are accumulated over an interval and then distributed as a batch.

The various rate coefficients are defined as follows:

♦ k_R (1/y) is the radioactive decay constant;

♦ k_{ij} (1/y) is the rate coefficient for transfer from compartment i to compartment j;

♦ k_{iL} (1/y) is the rate coefficient for loss from compartment L to outside the system.

Note that the model is normalised to a ground area of 1 m^2. Therefore, the animal component is the mass of animals associated with 1 m^2. This can be determined from the grazing density.

In respect of parameterization of the model, for the reference case it was assumed that $f_{fast} = 0.5$. For sensitivity studies, it was considered that f_{fast} should be varied on the interval [0, 1]. The amount of sludge or sludge product addition to agricultural land is typically 1 to 10 tons of dry solids per hectare. This corresponds to a value for M_{add} of 0.1 to 1.0 kg/m^2. The average rate recorded was 0.35 kg/m^2. Such additions might be made annually.

The parameter combinations $k_{13}+k_{14}$ and $k_{23}+k_{24}$ are the rapid and slow degradation rates, respectively. Thus, from Venter et al. [2001]:

$k_{13} + k_{14} =$ 55/y

$k_{23} + k_{24} =$ 1.1/y

For probabilistic studies, $k_{13} + k_{14}$ was taken to range from 25 to 85/y and $k_{23} + k_{24}$ was taken to range from 0.2 to 2.0/y.

In each case, degradation was assumed to lead to the production of $C(14)O_2$ dissolved in soil solution. This dissolved gas exchanges rapidly with carbon dioxide in the soil atmosphere. In practice, these exchanges are so rapid that these two compartments come almost instantaneously to equilibrium with similar molar concentrations in water and air. This is also the case for carbon dioxide in the below canopy atmosphere and carbon dioxide in the above canopy atmosphere. Therefore, the exchange rates between these various compartments can be set to large values in proportion to the mass of carbon dioxide in each compartment. More specifically:

$$k_{34}G_3 = k_{43}G_4 \qquad k_{47}G_4 = k_{74}G_7 \qquad k_{78}G_7 = k_{87}G_8$$

where G_3, G_4, G_7 and G_8 are the masses of carbon dioxide in compartments 3, 4, 7 and 8, respectively. These masses were estimated as described below. As noted above, all calculations are for a modelled area of 1 m^2.

The carbon dioxide concentration in the above-ground atmosphere is approximately 370 ppm on a volumetric basis. Taking the density of air to be 1.3 kg/m^3 and using the stoichiometric mass ratio of carbon in air of 12/28.8, the concentration is approximately 2 10^{-4} kg C/m^3. If the plant canopy is assumed to have an average height of 0.5 m, this means that G_7 is 1 10^{-4} kg C/m^2. It is noted that a rather smaller canopy height could be adopted for grazed pasture ($\sim$ 0.1 m), but that a value of around 0.5 m might be appropriate for hay fields and some agricultural crops. The height of the above-canopy atmosphere is less well defined. However, it will not be greater than the thickness of the atmospheric boundary layer. For the reference case, a value of 100 m was adopted, recognizing that the appropriate value in specific circumstances could be an order of magnitude larger or smaller than this. In particular, if the release area is limited in spatial extent, the degree of vertical dispersion will be much less than the total thickness of the atmospheric boundary layer and may be only $\sim$ 10 m. For a thickness of 100 m, the value of G_8 is 2 10^{-2} kg C/m^2. If the concentration of carbon dioxide in the soil atmosphere was the same as that in above-ground air, the amount present would be the product of this concentration, the depth of the soil zone (0.3 m) and the air-filled porosity ($\sim$0.1). This would give 6 10^{-6} kg C/m^2. However, because of microbial respiration, carbon dioxide concentrations in the soil atmosphere are more than one order of magnitude higher than those in the above-ground atmosphere. Therefore, G_4 was taken as 1 10^{-4} kg C/m^2, corresponding to a carbon dioxide concentration of 6200 ppm. The amount of CO_2 in soil solution, G_3, can be determined on the basis of equilibration with the soil atmosphere to be about 2 10^{-4} kg C/m^2.

The value of k_{43} was taken as 1 10^4/y, as internal exchange processes in soil are expected to occur on timescales of no more than hours. This then yielded $k_{34} = 5$ 10^3/y. Values of k_{47} and k_{78} were taken as 1 10^3, as these processes would be expected to operate on a timescale of days or less. This led to values of $k_{74} = 1$ 10^3/y and $k_{87} = 5$/y. It was noted that values of k_{47} and k_{78} are not well constrained.

The value of k_{8L} can be determined by considering the linear extent of the contaminated area and the wind speed across it. Assuming that the contaminated area has linear dimensions of 100 m and that a typical wind speed is about 5 m/s, an appropriate value for k_{8L} is $1 \ 10^6$/y (see also Section 5).

The uptake rate of carbon from the below-canopy atmosphere by plants will be governed by their requirements for energy production and growth. Taking the yield of pasture as around 1 kg (fresh weight)/m^2/y and assuming that the dry mass of pasture plants is 10% to 20% of their dry mass, leads to a dry mass production rate of 0.2 kg/m^2/y. This set a minimum on the carbon assimilation rate of about 0.08 kg C/m^2/y. To allow for the utilisation of assimilated carbon in respiration, a reference value of 0.2 kg C/m^2/y was adopted. With a mass of carbon in the below-canopy atmosphere of $1 \ 10^{-4}$ kg C/m^2, this led to $k_{75} = 2000$/y.

Conversely, the loss rate of carbon from plants due to respiration was taken as 0.12 kg C/m^2/y. With a standing biomass of plants of about 1 kg (fresh weight)/m^2, or 0.08 kg C/m^2, this led to a value of k_{57} of $0.12/0.08 = 1.5$/y.

The uptake of carbon from soil solution typically contributes no more than a few percent to the total carbon content of plants. A reference value of 1% was taken. As the amount of carbon obtained from the below-canopy atmosphere was defined to be 0.2 kg C/m^2/y, this led to the amount being obtained from soil solution being calculated as 0.002 kg C/m^2/y. As soil solution had a content of $2 \ 10^{-4}$ kg C/m^2, this led to a value for k_{35} of 10/y.

Internal transfers within plants were taken directly from the STAR model [Thorne et al., 2003b]. In their report, Thorne et al. [2003a] pointed out that $k_{56}G_5$ can be taken to be equal to $k_{65}G_6$, where G_5 and G_6 are the stable carbon contents of the fast and slow turnover compartments, respectively. On the basis of Smith et al. [1994], Thorne et al. [2003a] estimated that the stable carbon content of the fast turnover compartment is only about 0.2% of that of the slow turnover compartment. Thus, $k_{65} = 0.002k_{56}$. Furthermore, Thorne et al. [2003a] calculated $k_{56} = Z\varepsilon$, where Z is the turnover rate of carbon in the fast plant compartment (taken as 6/d from [Smith et al., 1994]) and ε is the assimilation efficiency (taken as 0.75 from [Smith et al., 1994]). This assimilation efficiency is somewhat higher than the value of 0.4 applied in the derivation of k_{75}. The lower value was used to derive $k_{56} = 2.4$/d or 880/y. This then implied that $k_{65} = 1.76$/y.

Loss rates from pasture by consumption by animals were estimated by dividing the sustainable yield of pasture (kg/m^2/y) by the standing biomass (kg/m^2). This approach was adopted by Thorne et al. [2003a]. They estimated a standing biomass of 0.5 to 2.0 kg (fresh weight)/m^2 and a yield of around 1 kg (fresh weight)/m^2/y. Thus, the loss rate from pasture was estimated in the range 0.5 to 2.0/y. Therefore, both k_{59} and k_{69} were set to 1/y.

The equations for animal compartments had to be normalised to the 1 m^2 of area used in the rest of the model. Beef cattle (mass 500 kg) typically consume 5 to 10 kg/d of dry matter [IAEA, 1994]. Consumption rates of dairy cattle are rather larger, at 10 to 25 kg/d [IAEA, 1994]. Taking an intermediate value of 12.5 kg/d and a dry matter content of 20%, the consumption rate was estimated at 62.5 kg (fresh weight)/d, corresponding to 22,800 kg/y. As discussed above, the yield of pasture is around 1 kg (fresh weight)/m^2/y. Therefore, the area required to sustain a single cow is approximately 22,800 m^2. As a typical cow has a mass of 500 kg, the mass of animal tissue associated with the 1 m^2 of model area is 500/22800 = 0.022 kg. This has to be taken into account when interpreting the model results.

Inhalation by animals was also included. As discussed above, it was estimated that a single cow grazes 22,800 m^2 of pasture. The total volume of above canopy air (to 100 m) in this region is 2.28 10^6 m^3. The breathing rate of a cow can reasonably be extrapolated from that of a man on the basis of metabolisable energy requirements. These scale approximately as body mass$^{0.75}$. As a reference man of mass 70 kg breathes about 20 m^3 of air per day, a cow of mass 500 kg will breathe about 90 m^3 of air per day or about 3 10^4 m^3/y. Thus, k_{89} = 3 10^4/2.28 10^6 = 0.013/y.

Values of the remaining coefficients were taken directly from the updated biokinetic model for animals set out in Thorne et al, [2003b]. The various values are listed in Table 3.

It should be noted that these various parameters are interrelated. This means that they cannot be varied independently over their ranges.

In summary, the rate coefficients adopted in the reference deterministic case are as set out in Table 4.

Table 3: Coefficients for the Animal Model for C-14

Parameter	Reference Value (1/y)	Range (1/y)
k_{910}	22.1	0 to 63.1
k_{109}	78.9	0 to 316
k_{911}	22.1	0 to 63.1
k_{119}	4.73	0 to 18.9
k_{912}	22.1	0 to 63.1
k_{129}	0.189	0.0631 to 0.631
k_{9L}	6310	3160 to 12600
k_{10L}	78.9	0 to 316
k_{11L}	4.73	0 to 18.9

Table 4: Reference Values of Parameters of the FSA C-14 Model

Parameter	Value (1/y)	Parameter	Value (1/y)
k_{13}	55	k_{87}	5
k_{23}	1.1	k_{89}	0.013
k_{34}	5000	k_{8L}	110^6
k_{35}	10	k_{910}	22.1
k_{43}	110^4	k_{911}	22.1
k_{47}	1000	k_{912}	22.1
k_{56}	880	k_{9L}	6310
k_{57}	1.5	k_{109}	78.9
k_{59}	1.0	k_{10L}	78.9
k_{65}	1.76	k_{119}	4.73
k_{69}	1.0	k_{11L}	4.73
k_{74}	1000	k_{129}	0.189
k_{75}	2000	k_R	0.00012
k_{78}	1000		

3.2   Illustrative Results

The C-14 model was implemented in AMBER and was run in both deterministic and probabilistic mode. The calculations were run for a continuous uniform input rate of 1 Bq/y to the agricultural environment for a period of ten years (by setting losses in the wastewater treatment plant to zero). Results of the deterministic calculations are shown in Figure 2.

Time (y)
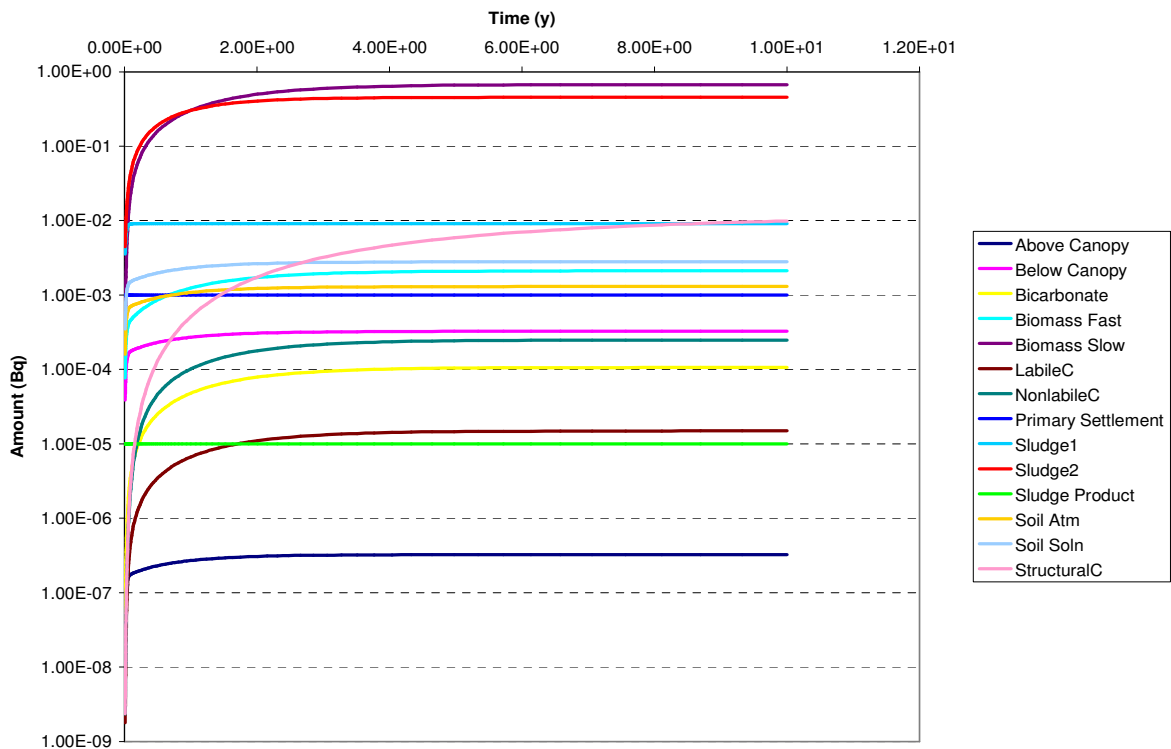
Figure 2: Results from Deterministic Calculations

For C-14, the characteristic timescale of the system is of the order of one year, except for Structural Carbon in animals, which continues to increase throughout the simulation period. Thus, apart from this compartment, results at ten years are representative of the equilibrium distribution of C-14 throughout this system for a continuous uniform input rate of 1 Bq/m^2/y.

Results at ten years are set out in Table 5.

The activities in the fast and slow degrading sludge compartments (Sludge 1 and Sludge 2) are found to build up to the expected equilibrium values. For the other compartments, their contents of carbon have already been specified or can readily be calculated. This allows specific activities to be determined. Results of this calculation are set out in Table 6 for a time of ten years after the beginning of the simulation.

The specific activities in soil solution and soil atmosphere are, as expected, very similar. That in the soil atmosphere is slightly lower, as there is some exchange with the below-canopy atmosphere, where the specific activity is further decreased by mixing with the above-canopy atmosphere. The specific activity in the above-canopy atmosphere is very low because of rapid dispersive mixing. The specific activity in plants is rather higher than in the below-canopy atmosphere. This reflects the fact that the rate of entry of carbon dioxide into plants for growth and energy requirements is greater than the rate of loss in respiration. This probably reflects a slight imbalance between the energetic value assumed for the carbon taken up and the biomass yield of the pasture that is cropped to provide a constant standing biomass. The total specific activity in animals is lower than that in plants, because much of the ingested carbon is exhaled rapidly as carbon dioxide. However, it is notable that the

contribution to the specific activity of structural carbon, which has a long residence time in the body, is tending toward the value calculated for ingested plant material.

Table 5: Results from the Deterministic Reference Calculation for C-14 at 10 Years

Compartment	Activity (Bq)
Sludge 1	9.09E-03
Sludge 2	4.54E-01
Soil Solution	2.79E-03
Soil Atmosphere	1.30E-03
Biomass (Fast)	2.11E-03
Biomass (Slow)	6.73E-01
Below-Canopy Atmosphere	3.25E-04
Above-Canopy Atmosphere	3.25E-07
Bicarbonate	1.06E-04
Labile Carbon	1.49E-05
Non-labile Carbon	2.49E-04
Structural Carbon	9.88E-03

Overall, the deterministic reference case performs as expected, except that plant concentrations of C-14 are possibly overestimated by a factor of two to three.

Basic model results are expressed in terms of amounts of activity in each of the model compartments. However, it is also convenient to report the results in terms of radionuclide concentrations. From the earlier discussion, it will be recalled that the C-14 model is formulated for 1 m^2 of soil. The soil depth modelled is 0.3 m, so the total mass of dry soil in the model compartment is 300 kg (based on a dry bulk density of surface soil of 1000 kg/m^3). The fresh mass of plants associated with 1 m^2 is taken as 1 kg and the mass of animals grazing 1 m^2, where appropriate, is taken as 0.022 kg.

Table 6: Specific Activity Values for C-14 for the Deterministic Case

Compartment	Activity (Bq)	Mass (kg C)	Specific activity (Bq/kg C)	Basis of Mass Estimate
Soil Solution	2.79E-03	2.00E-04	13.9E+00	
Soil Atmosphere	1.30E-03	1.00E-04	13.0E+00	
Biomass (Fast)	2.1E-03	1.60E-04	13.2E+00	Carbon content of plants of 0.08 kg C/m^2 partitioned as 0.2% to the fast compartment and 99.8% to the slow compartment.
Biomass (Slow)	6.73E-01	7.984E-02	8.42E+00	
Below-Canopy Atmosphere	3.25E-04	1.00E-04	3.25E+00	
Above-Canopy Atmosphere	3.25E-07	2.00E-02	1.63E-05	
Bicarbonate	1.06E-04	5.03E-03	2.11E-02	Animal mass of 0.022 kg/m^2, taking carbon to be 16/70 of the total body mass. Based on the carbon content and body mass of Reference Man [ICRP, 1975]. Note that the four specific activity values relate to the total mass of carbon and are, therefore, contributions to the overall specific activity of animal carbon.
Labile Carbon	1.49E-05		2.96E-03	
Non-labile Carbon	2.49E-04		4.95E-02	
Structural Carbon	9.88E-03		1.96E+00	

This allows soil, plant and animal concentrations to be defined as the sum of appropriate compartmental contents divided by the appropriate mass. The relevant compartments are defined as set out in Table 7.

It should be noted that the model calculation of concentration in soil neglects the soil atmosphere, as this would not normally be included in analyses of soil content.

Table 7: Basis for Calculation of Radionuclide Concentrations

Concentration	Formula
Soil	[Soil Solution]/Soil_Mass
Plant	([Biomass Fast]+[Biomass Slow])/Plant_Mass
Animal	([Bicarbonate]+[Labile Carbon]+[Nonlabile Carbon]+[Structural Carbon])/Animal_Mass

Detailed probabilistic results from the model are given in Thorne et al. [2003a]. The 95th percentile concentrations are typically about a factor of three higher than the 50th percentile concentrations.

4. CANADIAN STUDIES ON C-14

For both releases to atmosphere and to the soil zone, an important consideration is the degree to which methane is metabolized to carbon dioxide. In the post-closure radiological assessments undertaken by Nirex (e.g. [Nirex, 2003]), it has been assumed that methane is completely metabolized to carbon dioxide in the soil zone. This assumption is based on a review of the relevant literature carried out in 1991 [Stenhouse and Grogan, 1991].

In the context of discharges from CANDU reactors, Sheppard [2004] has studied the implications of releases of C-14 labelled methane. He notes that methane is not taken up by plants, but that it is ultimately oxidized to carbon dioxide both in soils and in the atmosphere. The following remarks and data are taken from his review.

Overall, about 7% (1-15%) of the methane released globally is oxidized in soil, whereas the majority is oxidized in the atmosphere by reaction with OH. These values are based on global averages. However, this does not imply that the oxidation in soil occurs close to the emission source.

Apart from the above estimates of global methane oxidation, there are various papers in the literature giving oxidation rates for atmospheric methane. Canadian researchers have been very active in this area so many of the data are directly relevant to assessment at Ontario power reactor sites. In Table 8, a summary of the oxidation rates of methane observed for sites relevant to Southern Ontario is provided. These data 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. These are represented by zero in Table 8.

There is a very distinct different in oxidation rate between forest and disturbed soils. Disturbed soils, such as agricultural soils have a much lower oxidation rate. Jensen and Olsen [1998] reported a 130-fold lower rate of oxidation in an arable soil compared with an adjacent forest soil. The geometric mean consumption rate from Table 8 is about 0.3 mg/m^2/d.

Table 8: Summary of Methane Oxidation Rates in Soils relevant to Ontario Power Reactor Sites [Sheppard, 2004].

Consumption (mg/m^2/d)	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]

5. DISCUSSION

In the context of the post-closure radiological safety assessment, the main gas of interest is C-14-labelled methane, although acetylene and ethylene may also be of some significance from Magnox wastes. The main factors that influence the radiological impact of C-14-labelled methane are:

◆ Area of release;

◆ Degree to which methane is metabolized to carbon dioxide in the soil zone;

◆ Uptake of C-14 by plants following metabolism of methane in the soil zone.

5.1 Area of Release

The area of release is primarily an issue of migration modelling, rather than biosphere impact estimation. It enters into the assessment calculations in that the concentration of C-14 in plants is inversely proportional to the loss rate from the above-canopy atmosphere. In turn, this loss rate is proportional to the square root of the release area and a geometrical factor that depends on the shape of the release area and on the distribution of wind speeds and directions at the site of interest. In the assessment calculations that have been undertaken by Nirex, the geometrical factor used has been that appropriate to a circular release area. With the near-uniform wind roses that occur in Britain, this is a reasonable approximation, as release over an extended narrow zone, as might occur along a fault, would be dispersed by winds blowing in a wide variety of directions relative to the long axis of the release area. The key uncertainty is in the release area, which could be as large (or larger) than the repository footprint, or could be restricted to a small area of faulting. However, it should be remembered that if the area is very small, it would not produce enough crops to sustain a reasonable potentially exposed group and dilution of the diet with uncontaminated foodstuffs would occur. It seems unreasonable to take an area of less than about one acre (4,000 m^2) or larger than the repository footprint area (about 300,000 m^2). With this uncertainty in area by a factor of 75, the uncertainty in plant concentrations due to this cause is about a factor of 9.

Although the area of gas release could be larger than the repository footprint area in some geological contexts, assumption of such a larger area for assessment purposes would be difficult to justify as a reasonably cautious approach.

5.2 Metabolism in the Soil Zone

The degree of metabolism in the soil zone is difficult to establish as experiments of direct relevance to this question do not appear to have been performed. However, Sheppard [2004] has estimated that the geometric mean consumption rate of methane in soils relevant to Ontario power reactor sites is 0.3 mg/m^2/d. 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 has been observed, with some indication that the lowest values occur in agricultural soils.

The maximum rate of methane generation from the repository in the Nirex generic performance assessment was calculated to be 5.05 10^3 m^3/y at STP. This corresponds to 3.6 10^6 g/y. If it is assumed that this is released over an area of 4,000

m^2, the flux is approximately 2500 mg/m^2/d. For a release area of 300,000 m^2, the flux is reduced by a factor of 75 to 33 mg/m^2/d. These values indicate that the highest fluxes of methane from the repository have the potential to overwhelm the metabolic capacity of methanogens in the soil zone, so reducing the quantity of C-14 metabolised to carbon dioxide. A cautious, but reasonable approach might be to calculate the specific activity of C-14-methane released from the repository and to then estimate the rate of C-14-labelled carbon dioxide produced in the soil zone by multiplying this specific activity by a mean consumption rate of about 0.3 mg/m^2/d. This would result in a reduction in maximum effective dose estimates from this pathway of a factor of 100 to 8,000, depending on the release area assumed. To put this in context, in the Nirex generic post-closure performance assessment, for the variant calculation in which the reaction of carbon dioxide with hydrogen was permitted, and for a release area of 10,000 m^2, the peak annual risk was assessed to be ~ 5 10^{-4}. Placing a limit on the rate of conversion of methane to carbon dioxide in the soil zone has the potential to reduce this risk to below the risk target. Rates of metabolism of acetylene and ethylene in the soil zone are likely to be very much less than rates of metabolism of methane.

5.3   Uptake by Plants and Animals

Finally, consideration should be given to the behaviour of C-14 after it has been incorporated in carbon dioxide in the soil zone. This is directly modelled in the C-14 model for sewage-sludge derived C-14 developed for the Food Standards Agency (FSA). In this model, an input rate of 1 Bq/m^2/y over a period of 10 years led to a specific activity in plants of 8.44 Bq/kg C (Table 6; summed over Biomass (Fast) and Biomass (Slow)) and a specific activity in animals of 2.04 Bq/kg C (Table 6; summed over all animal components). For comparison, the Nirex assessment uses the following regression-based formula to calculate concentrations in crops:

$$C_P = 1.13(M_{10}/8.84\ 10^{-4})^{-1}(\lambda_{1,9}/0.70)^{-0.4}(\lambda_{10}{}^{L}/1000)^{-1}$$

where C_P (Bq kg(C)$^{-1}$) is the concentration of C-14 in plants for a release rate to the root zone of 1 Bq/m^2/y;

M_{10} (kg/m^2) is the mass of stable carbon per unit area in the above-canopy atmosphere;

$\lambda_{1,9}$ (1/y) is the rate coefficient for transfer of carbon from plants to the below-canopy atmosphere;

$\lambda_{10}{}^{L}$ (1/y) is the loss rate from the above-canopy atmosphere.

The normalisation values for M_{10} and $\lambda_{1,9}$ in the equation for C_P are taken to be typical values of the associated parameters. Assuming these typical values, the expression reduces to:

$$C_P = 1130/\lambda_{10}{}^{L}.$$

The value of $\lambda_{10}{}^{L}$ is calculated explicitly using:

$$\lambda_{10}{}^{L} = n_S kv/A^{0.5}$$

where n_S is the number of seconds in a year (3.156 10^7);

k is a geometric factor relating to the contaminated area and takes a value of 0.752 for a circular area;

v (m/s) is the mean wind speed.

However, in the FSA model $\lambda_{10}{}^L$ corresponds directly to k_{8L}. For the reference calculation this is set to 1×10^6/y (Table 4). For comparison, for a release area of 10,000 m^2 and a mean wind speed of 5 m s^{-1}, the Nirex formula gives a $\lambda_{10}{}^L$ value of 1.19×10^6/y.

With $\lambda_{10}{}^L$ set at 1×10^6/y, the Nirex approach gives a specific activity in crops of 1.13×10^{-3} Bq/kg C. This value is much lower than that from the FSA model. Inspection of the FSA model shows that this arises from the assumed effectiveness of plant accumulation of C-14 from the sub-canopy atmosphere (compare rate coefficients k_{75} and k_{78} in Table 4.12). This further follows from the assumption that most plant carbon is derived from the sub-canopy atmosphere rather than the above-canopy atmosphere.

The Nirex model derives from studies with the detailed compartmental model RIMERS. The general structure of that model has been documented [Nirex, 1994] and is rather similar to the FSA model. Specifically, 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 do not appear to have been documented in any public domain literature. It seems likely that the substantial difference between the results reported relates 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.

5.4   Summary

In summary, in respect of C-14 releases in methane to soils, uncertainty in the area of release contributes about one order of magnitude uncertainty in the effective dose received. Assumption of complete metabolism of methane to carbon dioxide in the soil zone is thought to be highly cautious, because of the potential for saturation of microbial assimilation. Taking this into account could reduce effective dose estimates by a factor of 100 to 8,000, depending on the area of release. Conversely, the Nirex model may have significantly underestimated the degree of assimilation of C-14 by plants from the below-canopy atmosphere. Resolution of this issue would require detailed examination of the original data on which the model was based and computational comparisons with the FSA model.

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