BIOPROTA

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

Long-term Dose Assessment for Carbon-14

Report of an International Workshop

K Smith (Ed.)

Version 2.0
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PREFACE

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

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

This report describes presentations and discussions held during an international workshop on 28-30 April 2015, hosted by IRSN in Aix-en-Provence, France. Technical input provided by a wide range of organisations via presentations and discussions, as described in this report. The financial support provided for the workshop by IRSN (France), ENSI (Switzerland), Andra (France), EdF (France) SKB (Sweden), Posiva (Finland) and NUMO (Japan) is gratefully acknowledged.

Version History

Version 1.0: Draft workshop report prepared by Karen Smith (RadEcol Consulting Ltd) based on participant contributions and distributed to participants for comment on 24 June 2015.

Version 2.0: Final workshop report prepared, taking into account participant comments on the draft report. Distributed to participants and sponsors on 21 August 2015.

Executive Summary

Several collaborative activities concerned with long-term dose assessment for C-14 have been organised through the BIOPROTA Forum. These activities have contributed to greater confidence and reduced uncertainties in assessments made for disposal of solid radioactive waste containing C-14, such as graphite and reactor operating wastes. The most recent work was extensively reported in Modelling Approaches to C-14 in Soil-Plant Systems and in Aquatic Environments, published by the Swedish Radiation Safety Authority, as SSM report 2014:30, and in C-14: Data, Ecosystems and Dose Assessment, Report of an International Workshop, Aix-en-Provence, April 2014, available at www.bioprota.org. Results demonstrated the continuing potential for improved understanding of processes through collection, collation and sharing of new field and experimental data and its thoughtful application to assessment work. Such work could also benefit, and obtain support from, the assessment of doses due to current releases of C-14 to the environment. To address these developments and to share information on other recent C-14 assessment work, the current workshop was organised.

This report details presentations and discussions from that workshop. It was hosted by IRSN in Aix-en-Provence from 28-30 April 2015. The objective was to share information on data sets and application experience and to consider potential for collective interpretation and consideration in assessments.

The workshop was attended by 15 participants from 6 countries, representing a range of operators, regulators, researchers and technical support organisations.

A wide range of information was presented about the results of field work and experiments and their interpretation, other recent research, site investigations, model developments and approaches to building confidence in long-term C-14 dose assessments. Remaining uncertainties were identified and discussed, particularly with respect to C-14 in soil gas, including methane, carbon dioxide and carbon monoxide, and the role of larger organic molecules in longer-term retention and release. The role of carbon in ecosystems was highlighted, and the scope for using C-14 to better understand ecosystem behaviour and vice versa was recognised. The need to consider some short-term processes in order to understand longer-term equilibrium conditions was noted. Together, these issues highlight the potential value in sharing information arising from different research areas such as ecology, climate change, biogeochemistry and radiocarbon work, as well as radioactive waste disposal.

Suggestions for future activities included three items.

- Extension of model testing exercises to other modelling groups, based on comparison with monitoring data for atmospheric discharges collected by IRSN and results of field research and soil column experiments coupled with model development carried out for RWM.
- Application of research and monitoring information related to the Duke Swamp area at Chalk River Laboratories, AECL, Canada. These data may also provide a basis for testing the application of the geosphere-biosphere subsystem methodology developed through the BIOPROTA Forum.
- Review of the methods for characterising mires and understanding of carbon behaviour within different carbon pools, with a view to improving understanding the dynamics of transfers of C-14 among them, provide examples for site characterisation (what to look for, and how) and identify and define further model testing exercises.

CONTENTS

1. INTRODUCTION

Several collaborative activities concerned with long-term dose assessment for C-14 have been organised through the BIOPROTA Forum. These activities have contributed to greater confidence and reduced uncertainties in assessments made for disposal of solid radioactive waste containing C-14, such as graphite and reactor operating wastes. The most recent work was extensively reported in Modelling Approaches to C-14 in Soil-Plant Systems and in Aquatic Environments, Published by the Swedish Radiation Safety Authority, as SSM report 2014:30, and in C-14: Data, Ecosystems and Dose Assessment, Report of an International Workshop, Aix-en-Provence, April 2014[a]. Results demonstrated the continuing potential for improved understanding of processes through collection, collation and sharing of new field and experimental data and its thoughtful application to assessment work. Such work could also benefit, and obtain support from, the assessment of doses due to current releases of C-14 to the environment. Since the April 2014 workshop, new data and modelling work have been identified that may be available for collective interpretation and consideration in assessments. To address these developments and to share information on other recent C-14 assessment work, the current workshop was organised.

1.1 OBJECTIVES AND SCOPE OF THE WORKSHOP

The objective of the workshop was to share information, identified since the April 2014 workshop, relating to new data and model application experience and, as appropriate, develop ideas for future collaboration.

1.2 TECHNICAL AND FINANCIAL SUPPORT

The workshop was attended by 15 participants from 6 countries, representing a range of operators, regulators, researchers and technical support organisations. Participants and their organisations are listed in Appendix A.

Financial support for the workshop was provided by IRSN (France), ENSI (Switzerland), Andra (France), EdF (France) SKB (Sweden), Posiva (Finland) and NUMO (Japan). The technical and financial support of participants and sponsoring organisations is gratefully acknowledged.

1.3 REPORT STRUCTURE

Presentations from workshop participants and associated discussions are summarised in Section 2. Section 3 then provides an overview of the overall discussions and of possible collaborative tasks that could be taken forward.

[a] Available from www.bioprota.org.

2. PARTICIPANT PRESENTATIONS AND DISCUSSIONS

Following participant introductions, an overview of previous work and issues remaining was provided by the BIOPROTA Technical Secretariat. Presentations were made on developments in modelling approaches and assessments and potential data sets that could be made available for model testing. The presentations and associated discussions are summarised below.

2.1 REVIEW OF PREVIOUS WORK AND ISSUES STILL TO ADDRESS

Graham Smith (BIOPROTA Technical Secretariat) presented.

C-14 is naturally present in the environment, being generated in the atmosphere, but there are also a number of different anthropogenic C-14 source terms, including solid wastes (graphite, fuel components, reactor operating wastes and spent fuel etc.) and operational discharges from nuclear reactors, reprocessing plants, agricultural research and the pharmaceutical industry. C-14 can also enter the environment as a result of accidents. The different chemistry associated with these different wastes affects the source term. Information on the chemical form of carbon released from operating plants can be of interest in terms of providing information of relevance to biosphere assessments for waste disposal. As an example, boiling water reactors and pressurised water reactors have very different chemistries that will affect the C-14 source term and behaviour in the biosphere. Aside from the source term itself, there are many additional factors that will affect the chemical form of carbon in wastes that require disposal. Disposal of such wastes may be in near-surface facilities or deep underground; the decisions for which may be based on waste characteristics and/or the national regulatory regime.

C-14 is an issue that is not isolated to the nuclear industry. In the UK, at one time the largest assessed impact for a deep geological repository was associated with one waste stream from the pharmaceuticals industry (GE Healthcare Ltd), requiring the industry to address this issue. With a number of different industries and organisations being interested in C-14 in the biosphere, as a result of solid waste disposal and operational waste issues, assessment communities may have access to a range of sources of relevant information.

There have been some experiments undertaken on the speciation of carbon in wastes, such as graphite leaching experiments. However, it is generally considered in assessments that carbon is either present as carbon dioxide or not; measurements of speciation are generally not done, e.g. in leaching experiments evolved C-14 bearing gas that is not positively identified as carbon dioxide is tentatively considered to be methane, and this remains an important uncertainty in biosphere assessments.

The timeframes of interest for assessments varies. For solid waste disposal there is a need to consider the time prior to any release to the biosphere (as a result of near-field release and transport through the geosphere) and the duration of peak release. The timeframes will vary considerably depending on the facility, but after 20,000 years, radioactive decay will have been considerable and the importance of C-14 in assessments will reduce, although the quantity of C-14 in the facility will have an important influence on this. The period of peak release is often tens to hundreds of years, but may be longer for less degradable bulk wastes such as graphite. For operational discharges, timespans of hours to days are considerably more important with dominant releases occurring throughout the period of operation (tens of years). Although the timeframes of interest may vary, information on short term processes resulting from assessments and monitoring relating to operational discharges is required in order to understand the carbon pools present and the timescales over which they reach equilibrium.

Endpoints of interest in assessments can vary depending upon whether a cautious or realistic assessment is the objective. Assessments for people focus on individual dose and collective dose (distributed in time and space and by population group). Collective dose is not such a common endpoint currently, but is akin to the regulatory criteria for nuclear waste disposal in Finland and the respective approach taken by Posiva for 'other people' in their BSA-2012 assessment. This considered a wide range of exposure pathways, with intake of food and water being limited to the rates of the ICRP Reference Man in order to evaluate the spatial area of relevance. Within this area it was assumed that local produce was consumed before any externally supplied produce, taking into account the food productivity of the land and waters within the assessment area. Due to the low productivity of the area, water consumption was the dominant pathway, with the peak dose occurring at 5,000 years post closure in the base scenario, the majority of the dose being explained by C-14 in ingested water.

In addition to human dose assessment endpoints, there is an increasing requirement to consider dose rates to populations of non-human species. For longer assessment timeframes, beyond those that can reasonably be evaluated in terms of dose assessment, concentrations into and fluxes through environmental media can be an endpoint of interest. It was noted that within the EU COMET project there is increased interest in other measures such as epigenetic changes, genomic instability and other alternations in biochemical, cytological and physiological characteristics as measures of the adverse impacts of ionising radiation on organisms. It was further noted that, for C-14 in organic form, there may be a need to consider how to address impact following incorporation of this low-energy beta emitter into biota and specifically into particular cellular organelles, DNA and RNA. With the focus of biota assessments being on populations, there is a need to consider the spatial and temporal distribution of radionuclides and exposed populations, which is similar to considerations for collective doses for human populations. Carbon is an essential element and a building block for all life forms. As such, a good understanding of an assessment is needed, along with a system description, to allow C-14 dynamics to be evaluated.

Whilst there has been a great deal of effort spent on addressing C-14 dose assessment, the issue keeps returning. In 2005, BIOPROTA published a first report on C-14, focussing on reviewing the different models available for assessing C-14 doses to people and more detailed work has been undertaken since this initial report:

- Limer L, Albrecht A, Marang L, Smith K, Thorne M C, Wiebert A, Xu S and Smith G (2009). C-14 Long-Term Dose Assessment: Quantitative Model Comparison and Development, Part I. www.bioprota.org.

- Limer L M C, Smith K, Albrecht A, Marang L, Norris S, Smith G M, Thorne M C and Xu S (2011). C-14 Long-Term Dose Assessment in a Terrestrial Agricultural Ecosystem: FEP Analysis, Scenario Development, and Model Comparison Published by the Swedish Radiation Safety Authority, as SSM report 2012:47.

- BIOPROTA (2013). Modelling Approaches to C-14 in Soil-Plant Systems and in Aquatic Environments. Report of a review and International Workshop held 12 – 14 February 2013. V3 draft final issued July 2013; SSM report 2014:30.

- BIOPROTA (2014). C-14: Data, Ecosystems and Dose Assessment. Report of an International Workshop, 1 – 3 April 2014, Aix-en-Provence. www.bioprota.org.

Journal articles have also been produced and international conferences attended to develop interaction with other C-14 assessment communities[b].

Since the last BIOPROTA C-14 report in 2014, the US NRC has released a phase 2 report[c] on the analysis of cancer risks in human populations near nuclear facilities that sets out general methodological considerations for health and dosimetry data processing, including recommendations on dose assessments. Within those recommendations it is noted that particular attention should be paid to the releases of carbon-14, which is believed to account for a substantial fraction of the doses in recent years and yet modelling capacity was considered to be relatively weak. The focus here is not on demonstrating compliance with some limit, but rather on assessing risks. As such, conservative screening models and approaches are not appropriate; process orientated models are required. The science required to support such models is very much aligned to what has been, and continues to be, considered within BIOPROTA.

There are notable questions that arise in relation to the ICRP dose coefficients for C-14 in drinking water. 100% absorption and utillisations is a reasonable assumption for C-14 in food, but for drinking water it may not be so reasonable, e.g. for C-14 present as dissolved CO_2, bicarbonate or carbonate. Inhaled carbon dioxide diffusively exchanges across the lung effectively so assuming 100% absorption here is reasonable; however, only a small proportion of that absorbed is then utilised, the majority exchanges back across the lung and is exhaled. If C-14 in drinking water is in the form of carbon dioxide, this will be absorbed into the bloodstream and be lost via the lungs rather than being utilised to any great extent in metabolic processes. Dissolved bicarbonate or carbonate may also not behave in the same way as carbon in food. Advice on appropriate dose coefficients for C-14 as taken into the body as dissolved CO_2, bicarbonate and carbonate has been requested from ICRP Committee 4[d].

Special approaches to modelling are required for C-14 and two broad types are commonly applied. Specific activity models are applied in many contexts, but consideration needs to be given to the different carbon pools and whether it is reasonable, in the context of the assessment being undertaken, to assume that they are in equilibrium. Process orientated models are also applied, but require a good understanding of the system being modelled in order to be able to characterise processes sufficiently. Both approaches therefore require a good understanding of the ecosystem.

Although various C-14 specific models are available, C-14 remains a large issue in dose assessments and further improving the confidence in assessment results is therefore worthwhile. Continuing issues relating to C-14 models include:

- The dynamics and chemical forms of different source terms;
- Dose coefficients for C-14;

[b] For example, see: Mobbs S., Shaw G., Norris S., Marang L., Sumerling T., Albrecht A., Xu S., Thorne M., Limer L., Smith K. and Smith G. (2013). Intercomparison of Models of ^{14}C in the Biosphere for Solid Radioactive Waste Disposal. Proceedings of the 21st International Radiocarbon Conference. Radiocarbon 55 (2-3): 814-825. The next international Radiocarbon Conference will take place in November 2015. BIOPROTA representation at this conference may be of merit.

[c] Report available at www.nap.edu.

[d] Similarly, advice has also been sought from ICRP concerning the appropriate dose coefficient for Cl-36 when consumed by an individual who also consumes the typical intake of salt, as opposed to the amount needed.

- Methane oxidation and emanation from soils; and
- Carbon pools and fluxes in aquatic systems.

An important mechanism by which confidence in assessment models could be improved would be to identify monitoring data that could be applied to support the application of assessment models and provide the means by which model-data validation could be undertaken both for terrestrial and aquatic releases. Further benefits could be gained by bringing together the modelling communities interested in solid waste disposal with those interested in current releases, since models and system understanding interests are not considered to be particularly different. There may also be scope to learn from the geo-storage community and others, including peatland scientists and those involved in climate assessments. Most solid waste disposal assessments consider a first pass of carbon through a system, whereas climate change work relating to carbon takes greater account of carbon cycling and associated timescales. Information from this assessment group could be very interesting and would help considerably in developing our understanding of carbon cycling in soils and mires (as well as in marine systems). There is also scope to raise ecology-related issues with the recently established IUR FORUM.

The need to work through the realistic versus cautious modelling approach issue in relation to the appropriate level of model required was highlighted; an actual assessment may well require a mix of both approaches to ensure a balanced level of detail in the context of the assessment question.

Discussion

Where releases to the biosphere are in the form of carbon dioxide, it may be possible to undertake a comparison with monitoring results following weapons testing etc. Whilst information on mechanisms may be lacking, it would be possible to evaluate whether impacts are below those resulting from previous releases to the biosphere. The difficulty arises however, when different forms of carbon are released or where specific biochemical processes lead to concentration in the biosphere or in cellular structures. Biological mechanisms can vary and this should be taken into account.

There is the potential to learn from work undertaken by others in relation to C-12/C-13 ratios, which could help to develop understanding of how carbon signals are propagated throughout the environment. C-14 itself is difficult to measure in situ and, whilst there may be a different isotopic behaviour associated with a mass of C-14, it is unlikely that this would equate to more than a few percent discrimination. There are nonetheless unresolved questions relating to carbon isotopic ratios.

Increased concentrations of carbon dioxide can lead to enhanced plant productivity. However, if too much carbon dioxide is present, this can prove toxic to plants. Natural analogue experiments have previously been undertaken as part of the FACE (free-air carbon dioxide enrichment) experimental programme, where transects around natural venting areas were studied and changes in the growth and population structure of natural vegetation were observed. Results indicate that high levels of carbon dioxide can affect ecosystems. Such impacts may therefore need to be considered in assessments where bulk gas release may occur from a repository. The potential for increased carbon dioxide to increase soil acidity with subsequent impacts on plants via roots may also warrant consideration. It is not only carbon dioxide that needs to be considered in relation to plants however, they are also known to take up formaldehyde and other organic forms, extracting carbon dioxide from these for use in metabolic processes and biomass production.

Consideration of a wider group of people than a common ‘reference group’, such as considered by Posiva in their BSA-2012 assessment was suggested as a possible recommendation to the assessment community. The productivity of land was noted as an important consideration and should be considered in combination with realistic assumptions around human behaviour and the area of land contaminated as a result of the passage of C-14 through the geosphere-biosphere interface. This subject area is due to be further discussed during a ‘continuing issues’ BIOPROTA workshop to be held in May 2015, relating to a potential update of the IAEA BIOMASS reference biospheres approach. The use of appropriate models adapted to different radionuclides within a common calculational framework will also be a subject of discussion within this proposed programme.

From discussion, it was noted that it would be helpful to have a set of physiological models for nutrient elements for which there are relevant radioisotopes, e.g. P-32, P-33, Cl-36, Se-79, I-129 as well as C-14.

2.2 ASSESSMENT OF C-14 IN FRESHWATER LAKE FISH

Achim Albrecht presented.

C-14 is acknowledged as a particular issue for a low level waste (LLW) surface-based repository in France. There are diffusive losses of C-14 from waste containers plus atmospheric releases from waste treatment plants. Currently, C-14 is only considered in the form of carbon dioxide.

Atmospheric models for evaluating the transport of gaseous emissions and wind roses are available for the region that allow key areas of interest, in terms of the atmospheric plume, to be identified. Small lakes have been identified as a result. Planned emissions equate to a volumetric concentration of 7E-07 Bq/m^3 C-14, which in itself does not drive toward the development of a fish model for C-14 in lakes, but such a model would be required in the case of accident situations.

The Andra model uses a specific activity approach and, as such, there is no requirement to consider specifically the lake water concentration of C-14 when the atmospheric concentration is known, as fish concentrations can be linked directly to the atmospheric concentration. C-14 deposition is not a required process as carbon dioxide does not deposit, although some interaction with rain drops may occur. Local information on the consumption of fish in the area is currently being gathered to inform the dose calculations.

Should organic forms of carbon be present then the situation is more complex. Atmospheric deposition, that can be ignored for inorganic C-14 must be considered for organic species. Furthermore, there seems to be evidence that organic forms of carbon are preferentially taken up by fish and relevant information on uptake by fish of different organic substances may be gained from fish toxicology studies.

Release of C-14 to atmosphere from the facility will be low throughout its circa 50 year operational period. There may also be some discharge to groundwater that may subsequently enter surface waters. However, monitoring to date has indicted that there are no measureable concentrations of C-14 in groundwater arising from the facility. There is nonetheless the possibility that organic forms of carbon may occur in groundwater in the future and consumption by fish therefore requires consideration.

Discussion

The isotopic ratio of C-14 in air controls that within water and in fish, hence the argument has been made that the model does not need to specifically include sediment. It is understood that a further step has been taken previously by Steve Sheppard with regard to chlorine whereby it is assumed that the isotopic ratio in people is equivalent to that in air. In the case of the French lake scenario, the only source of contamination is from atmospheric exchange (for C-14 in the form of carbon dioxide). It was noted however that C-14 would be assimilated in the terrestrial environment, leading to a change in the isotopic ratio and this flux, over the longer term, would be transferred to the lake via dissolved organic matter. Interactions between air, the terrestrial environment and the lake may therefore be important; with the lake being situated some 3 to 4 km from the site, only a low C-14 concentration would occur in water as a result of direct atmospheric exchange, but release and uptake throughout the entire catchment area with uptake to plants could result in concentrations in the lake that could potentially be an order of magnitude higher. The main flux of C-14 to the lake will therefore be from the terrestrial environment. It was furthermore noted that the turnover of water in the lake should be considered and the source of water entering the lake. Recent evidence suggests that most of the water that seeps through a soil into surface water bodies carries carbon that is not accumulated in soils.

The SKB approach to C-14 has been to construct an overall carbon balance of the system and then to consider the C-14 inventory and distribute this in proportion throughout that system. The issue then is primarily on evaluating the C-14 inventory in the waste.

2.3 C-14 DOSE ASSESSMENT IN SR-PSU

Peter Saetre presented.

SFR is an operational facility for the disposal of low and intermediate level waste at Forsmark in Sweden. An application to extend the facility, as depicted in Figure 1, has recently been submitted by SKB to the Swedish regulator, SSM. The application is supported by the safety assessment SR-PSU, which concluded that the extended SFR repository would meet regulatory criteria with respect to long-term safety.

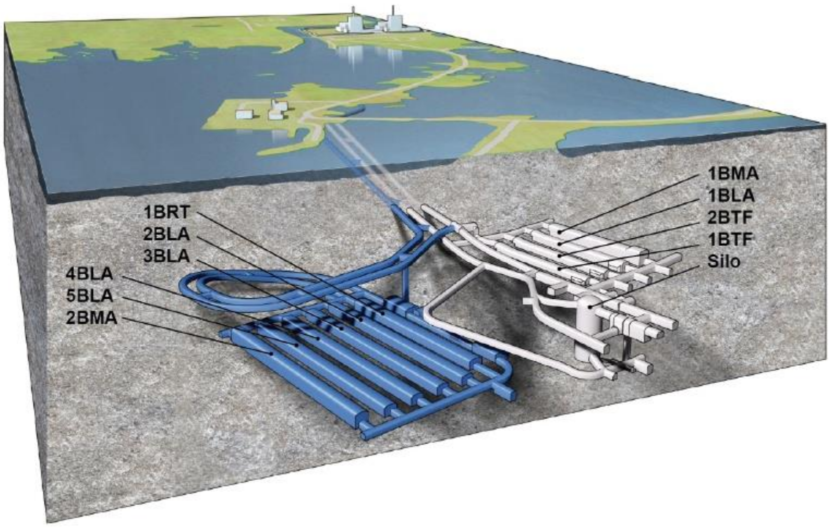

Figure 1. The SFR disposal facility for low and intermediate level waste at Forsmark illustrating the current operational facility (white) and the planned extension (blue).

A range of different scenarios was considered in SR-PSU, but the focus of the presentation was on the main scenario.

C-14 was expected to be an issue and a radionuclide specific model was therefore developed. The model was intended to evaluate whether C-14 was indeed an issue and, if so, to allow important features and processes to be identified.

The highest doses to people were associated with a wetland where radionuclides can accumulate and that is subsequently drained and used for agriculture. The key radionuclides contributing to dose over time were C-14, Mo-93 and then Ni-63 toward the end of the assessment period (Figure 2).

The key crops contributing to doses were tubers, fodder and cereals. Differences in C-14 content of crops were due to differences in plant canopy dynamics. The model demonstrated that approximately 50% of C-14 is taken up by the roots in the form of carbon dioxide dissolved in soil water. This equates to a 2% uptake of total carbon by roots, but, as a result of the higher C-14 to C-12 ratio in soils as compared with air, this gives rise to 50% uptake of C-14. The carbon dissolved in soil water is of greater importance for plant uptake than organic carbon in the soil. Important considerations are therefore the soil diffusivity coefficient and concentration of the stable inorganic carbon dissolved in soil water.

The greatest C-14 concentration in the lake occurs just prior to the lake becoming fully isolated. Fish concentrations are therefore greatest at this time and fish consumption by local people was therefore an important parameter. Reasonable constraints were applied however to avoid unrealistic assumptions about the quantity of contaminated fish that can be consumed by a single person.

Within SR-PSU it was necessary to consider uncertainties around future climate. The base case allowed for global warming in line with elevated carbon dioxide levels of a similar magnitude to the present day. An extended warming scenario was also considered that gives rise to a large-scale increase in seawater levels. The extended warming also gives rise to a wetter climate and an increase in the flux of surface and groundwater.

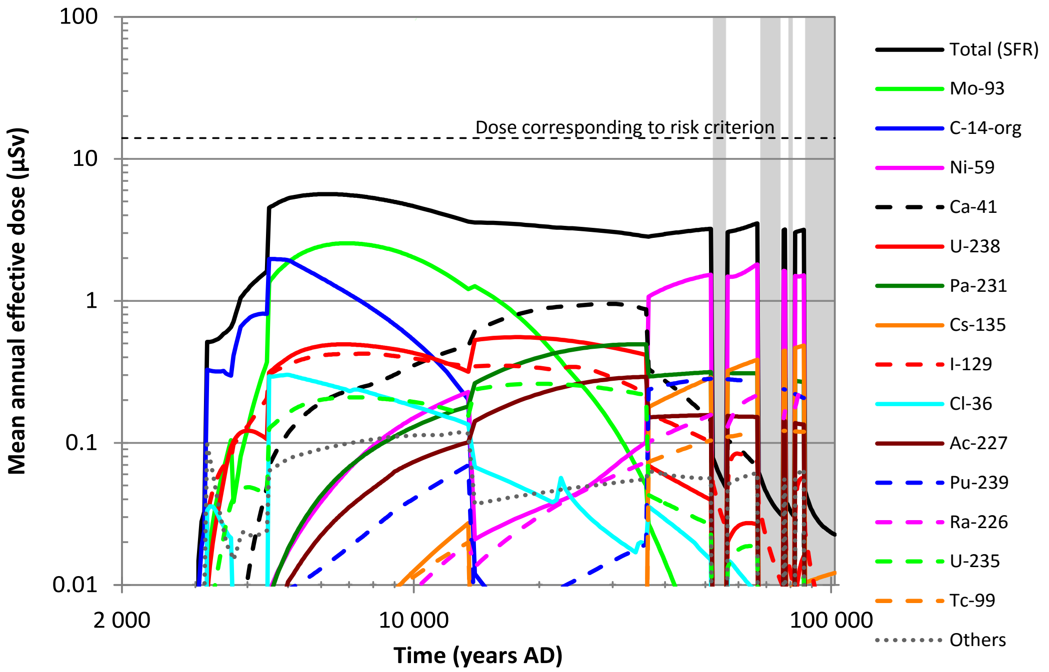

Figure 2. Key radionuclides over time in the main scenario of the SR-PSU safety assessment.

For a periglacial climate, the depth of permafrost can be modelled, but it is harder to model where taliks will occur. Building on hydrological modelling of permafrost conditions at the site, it was assumed that radionuclides from the repository will reach topographical lows above the repository or a large lake further away. Permafrost prevents cultivation and limits the ability to extract well-water. Consequently, the assessment focuses on a hunter-gatherer lifestyle in a low productivity system.

A colder climate was also considered to evaluate the effects of glaciation on the site (although this calculation case was not part of the main scenario). Once a glacier has receded, the site would be inundated with water due to depression of the land. Land evolution is an important component of the assessment therefore. Release of radionuclides in groundwater to the biosphere could occur to a coastal sea basin, a wetland or a lake depending upon the timing of the release. These release areas are not co-located.

Of the climate cases evaluated, the base case (global warming) gave rise to the greatest doses although the doses calculated for the extended warming case were of a similar order of magnitude. Doses arising from glaciation and permafrost scenarios were both lower than in the base case.

In their feedback on a previous assessment, SSM has asked about the size of biosphere objects and how certain it is that discharges will occur to one of them. Some judgement is required in delineating biosphere objects and alternative object delineation has therefore been considered. For the SR-PSU assessment, information was available on release points to the biosphere from particle tracking model simulations and this was used to identify the key biosphere objects where releases may occur. One alternative discharge area was identified from the density of particles at the geosphere-biosphere interface, and another was identified based on an assumed continuous flow of groundwater upward from a depth of 60 m. Open wetland areas were also considered due to concerns over rare frogs and vascular plants, and these were evaluated in terms of impacts on non-human biota.

For each alternative biosphere object, a unique description of hydrological fluxes and regolith depths were derived from three dimensional representations of the area. For the transport simulations it was then assumed that a constant release of radionuclides reached the alternative biosphere objects evenly.

For a scenario where a well is constructed in the lowest soil layer, C-14 concentrations increase as the biosphere object decreases in size, which is a function of groundwater flow. C-14 concentrations in surface peat pore water also increase as a function of size. In this case, loss of C-14 from peat is via degassing, which is proportional to the soil surface area. The consumption of fish from a downstream lake is an important pathway for C-14 doses. If the catchment area of the upstream objects is reduced, the flow of water into the downstream lake is also reduced. However, C-14 concentration in the groundwater exported to the lake is highly dependent on the rate of degassing (see above), and thus the net effect of object area was that C-14 concentrations in lake water tended to increase with decreasing object size. Though alternative outlines of the primary biosphere objects had a clear effect on environmental concentrations, the effect was limited. That is, the concentrations in outlined areas most likely to receive contaminated groundwater were typically not more than two or three times larger than the concentration in the original object.

The C-14 model has been checked using a C-12 model. Carbon sources include inorganic carbon from the atmosphere, inorganic carbon from deep groundwater, and inorganic and organic carbon (that can partly degrade to inorganic forms) from surface run off and stream water. The dissolved inorganic carbon (DIC) model results were found to be similar to empirical estimates for lakes. This was also similar for a wetland although the initial degassing rates were adjusted to improve the accuracy of the model in terms of pore water DIC. Younger mires accumulate carbon faster than older mires and results for long-term carbon accumulation in peat were found to be similar to measured data.

One issue in developing models is that, at the point of model development, the biosphere objects of most importance for dose calculations are not known. This tends to drive toward larger, more complex models. Input to biosphere model development is also required from near-field and geosphere modelling. Once important areas have been identified, it is then possible to simplify the model.

Discussion

Degassing rates could be greater than assumed in the model if groundwater is at greater pressure when entering the surface, leading to greater outgassing. The influence of groundwater pressure on degassing rates has not yet been considered.

Results from the biosphere modelling are primarily presented in TR-14-16 (Chapter 10)[e]. Further information on biosphere parameters, including root uptake, is provided in SKB report R-13-18. The radionuclide model report R-13-46 also provides useful information on the approach taken to evaluating C-14 in the SR-PSU assessment.

2.4 HOW TO CONSIDER THE DIFFERENT BEHAVIOUR OF INORGANIC AND ORGANIC CARBON IN DOSE ASSESSMENT OF C-14? MORE QUESTIONS THAN ANSWERS!

Achim Albrecht presented.

Inorganic and organic forms of carbon have significantly different behaviour. Organic forms, such as methane, are reduced forms of carbon, whereas inorganic forms such as carbon dioxide are oxidised forms. A distinction may also be warranted between aliphatic and aromatic forms. Inorganic forms are likely to react with concrete and can exchange with carbonates present in altered concrete and host rock. Organic forms such as methane are less likely to react and are thus less retarded. Transport in the gaseous or dissolved phase is therefore possible, although methane has a much lower solubility than carbon dioxide. No general statement can be made for more complex organic substances.

The formation of organic substances at room temperature requires microbial or enzymatic catalysis. Leaf tissues of living plants emit methane, whereas humans and animals release methane from the gut. The oxidation of organic substances back to carbon dioxide again requires microbial (enzymatic) catalysis. Organic matter can be subdivided into readily bioavailable forms such as sugar/cellulose, and more persistent forms such as soil humus and persistent organic pollutants.

Redox reactions within organic material give rise to Gibbs free energy. The energy released from the oxidation of methane is variable depending upon the redox reaction. Humic substances in soil are normally thought of as non-reactive, but fungi, in the absence of more readily available carbon, can break down these complex forms. Microbes can also break down these forms, but again more available forms of carbon must be absent.

The chemical degradation of cellulose in concrete is the relatively well known under alkaline, anaerobic conditions that would be expected in the near field, giving rise to isomers of isosaccharinic acid (ISA). Some of the resultant isomers can complex other radionuclides such as plutonium. There is a need therefore to consider what is expected to happen in the near field and, from this, determine which organic substances are likely to be present that would be expected to reach the biosphere.

EdF is currently dismantling an early graphite reactor. To evaluate the chemical forms of carbon that can result from graphite waste, graphite was placed in 0.1M NaOH solution. Results indicate that the majority of the forms extracted were inorganic (Figure 3), but around 30% of forms were organic. Both

[e] See www.skb.se/publications

forms have therefore had to be considered. More complex organic forms such as cellulose, polymers, phthalates etc. may be associated with low to intermediate level wastes.

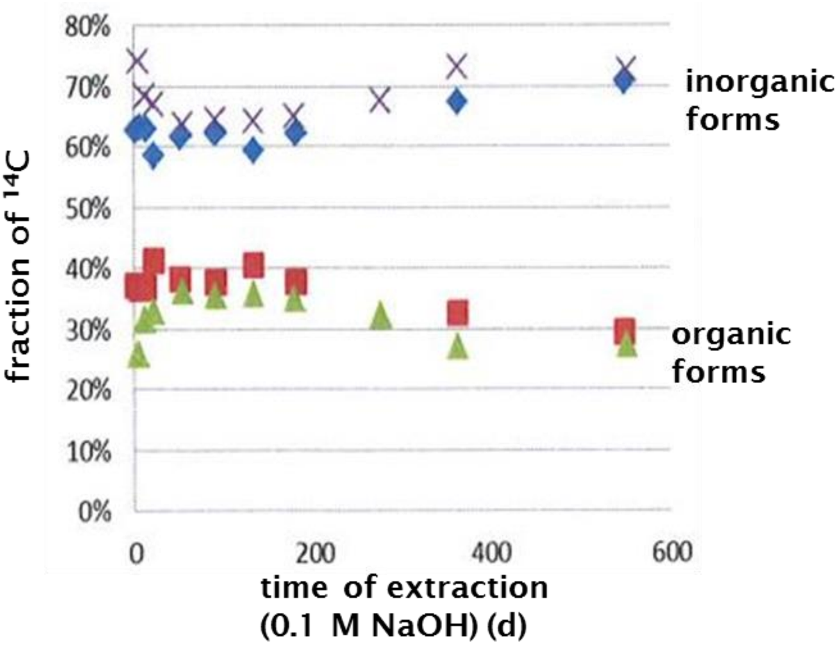

Figure 3. Extraction of inorganic and organic forms of carbon from graphite waste by sodium hydroxide.

It may be possible to gain knowledge from landfill sites such as the Oulens landfill facility in Switzerland, which is now closed. Whilst the chemistry inside the facility is unknown, polymer covers and sand-bentonite mixtures are in place that are similar to the barriers in place at LLW facilities. Landfill gas production measurements could provide useful information on the forms of carbon that can pass through such barriers. The production of landfill gas over time is fairly well understood. Oxygen present in the facility initially is rapidly reduced with carbon dioxide being produced. Once oxygen is fully utilised, methane begins to be produced in approximately equmolar quantities with carbon dioxide. Whilst the theory behind the generation of the different carbon gasses is well understood, there is a need to more fully understand the components, kinetics and thermodynamics of the processes in order to evaluate what happens to the different forms of organic matter within a facility and the timeframe associated with the different reactions and release of different gasses.

The classical view of the behaviour of methane and carbon dioxide in soils is illustrated in a publication by Carmichael et al (2014)[f]. The view is that there is always within a soil, an area where there is no oxygen input, which allows for methanogenesis. Methane production may be from fermentation or the reduction of carbon dioxide to methane. Methane can be transported through plants or can accumulate in soils and lakes etc. with ebullition to the atmosphere. Alternatively, methane can be re-oxidised as it moves into aerobic soils. A more contemporary view takes account the role of vegetation in the cycling of methane and other important factors such as climate. The phloem of plants can transport methane

[f] Figure 1 in Carmichael, M.J., E.S. Bernhardt, S.L. Bräuer and W.K. Smith (2014). The role of vegetation in methane flux to the atmosphere: should vegetation be included as a distinct category in the global methane budget? Biogeochemistry 119:1-24.

even though it is not utilised and this transport pathway for methane may need to be considered when evaluating the amount of methane that is being oxidised to carbon dioxide in soils.

There is a wealth of information available on methane in the biosphere, largely as a result of global warming-related research (methane is important in terms of climate change, being a more potent though less persistent greenhouse gas than carbon dioxide), but consideration needs to be given as to how this information should best be used in assessment models. Around half of the global soil carbon pool is in the arctic region. As permafrost conditions recede, unstable ground conditions could form as land areas defrost, which could give rise to the sudden release of methane, formed as a result of organic matter decomposition.

If methane is formed in soils and diffuses upward, consideration must be given to its rate of conversion to carbon dioxide. If methane is not converted, it will be lost to air. Where conversion to carbon dioxide occurs, this may be taken up by plant roots or through the plant canopy following release from the soil surface. In higher carbon dioxide atmospheric conditions, plants tend to grow faster and more carbon can therefore be retained in organic matter; there is a close coupling between carbon dioxide concentrations and plant productivity.

The turnover of organic matter in soils is not well understood, particularly in relation to complex organic matter. Priming of soils is important for agronomists whereby soil additions are made to enhance the value of the soil. Such additions lead to a greater release of carbon and other nutrients required by plants. When organic matter is added to soils, a higher yield occurs in first or second years post-addition. The yield then drops. By adding organic matter to soil, the agricultural yield is enhanced, but the addition gives rise to a loss of organic matter over time and a resultant loss in sustainability. The cycling of organic matter in soil is still a subject under discussion within the scientific community and the long-term assessment community needs to consider what data we require and under what conditions.

Schaffer et al [2015] undertook a simple experiment on the influence of a compost layer on the attenuation of micro-organic-pollutants[g]. The experiment aimed to investigate whether organic substances were degraded or retarded etc. Results indicate a wide range of behaviours for the different micro pollutants. The behaviour of organic substances is not therefore a simple question. Microbial activity is very important and this is controlled by carbon and other nutrient availability and soil redox potential, the solubility of trace metals, temperature and soil hydrology. Many of these factors are strongly coupled and some are affected by soil pH. The coupling between parameters therefore requires consideration as many factors influence others.

Bishop et al (2015)[h] looked at carbon isotopic ratios in different catchment areas. Results indicate that the carbon found in streams is largely modern. This suggests that it may not be necessary to consider ‘old’ carbon in long-term assessments.

Whilst the research undertaken to date has led to a large database with associated empirical data and modelling, there is still no consensus. A statistical approach to evaluating and selecting from the available data for use in long term assessments may be required. As a basis, structuring all of the

[g] Schaffer et al. (2015). Influence of a compost layer on the attenuation of 28 selected organic micropollutants under realistic soil aquifer treatment conditions: insights from a large scale column experiment. Water Research 74: 110-121.

[h] Bishop et al. (2015). Getting terrestrial carbon into the aquatic conduit: Riparian peat controls from daily to centennial time-scales. Geophysical Research Abstracts 17.

available information may be beneficial. Not only would this provide a useful resource for further developing our understanding of key processes and selecting appropriate parameter values, remaining knowledge gaps and uncertainties would be more readily identifiable.

Discussion

Formaldehyde has been shown to be accumulated in microbial biomass, and can be further processed to carbon dioxide. A delay between methane injection into soils and carbon dioxide production was evident from experiments conducted on behalf of RWM (Radioactive Waste Management Ltd, see section 2.5 below). In addition to carbon dioxide, carbon monoxide can also be generated. It is easy for assessors to get fixed on photosynthesis as the means by way C-14 enters into plants, but carbon monoxide assimilated via serine could be an alternative pathway for C-14 incorporation into living plant biomass. Carbon monoxide is measured in graphite release and it would be interesting to see route it goes down, whether carbon dioxide or methane is formed on entry to the biosphere (or potentially as a result of microbial processes in the near-field).

There has been a lot of work done historically on microbial activity in the near-field and both experimental and modelling work has been undertaken to evaluate microbial activity in waste. In a host rock comprised of clay, it may be possible for sulphate to leach into the near field. Sulphate is a preferred energy source for microbes. Where sulphate is present, methane production may not occur.

2.5 TRANSPORT OF C-14 IN AGRICULTURAL ECOSYSTEMS: RESULTS FROM LABORATORY AND FIELD INVESTIGATIONS

Mike Thorne presented.

There are many different natural processes that can affect carbon cycling in the biosphere that act on different timescales (Figure 4). The interplay between lateral and vertical transport of carbon is determined by the source term and whether this is from above or below. Turbulence within a plant canopy affects the upward and downward cycling of carbon and also the lateral transport through the canopy. The transport of gas and water through plants varies considerably in time with gas transport being rapid (seconds to minutes) and water transport much slower (minutes to hours). The interplay between photosynthetic processes, respiration and growth occurs over timescales of hours to seasons. The return of organic matter to soils occurs over timescales of seasons to millennia, depending upon the organic form.

Microbial oxidation/reduction of organic matter can occur in a matter of seconds to minutes, but this very much depends on the carbon source. Uptake of carbon by plant roots could occur via either passive or active mechanisms. Movement in the transpiration stream can then occur. Gas transport through plant aerenchyma can also occur. The various processes take place under varying hydrological conditions that can themselves vary in timescales ranging from minutes to millennia (e.g. associated with global climate change).

Where a source term is an atmospheric discharge, input of C-14 to the system could be constant over time for routine releases, alternatively there could be rapid release associated with non-planned events. For source terms associated with geosphere release to the biosphere, the timescales can be highly variable. Timescales are routinely thought of as long-term (years to millennia), but more rapid release could occur, for example the accumulation of gas in a geological trap with subsequent borehole penetration.

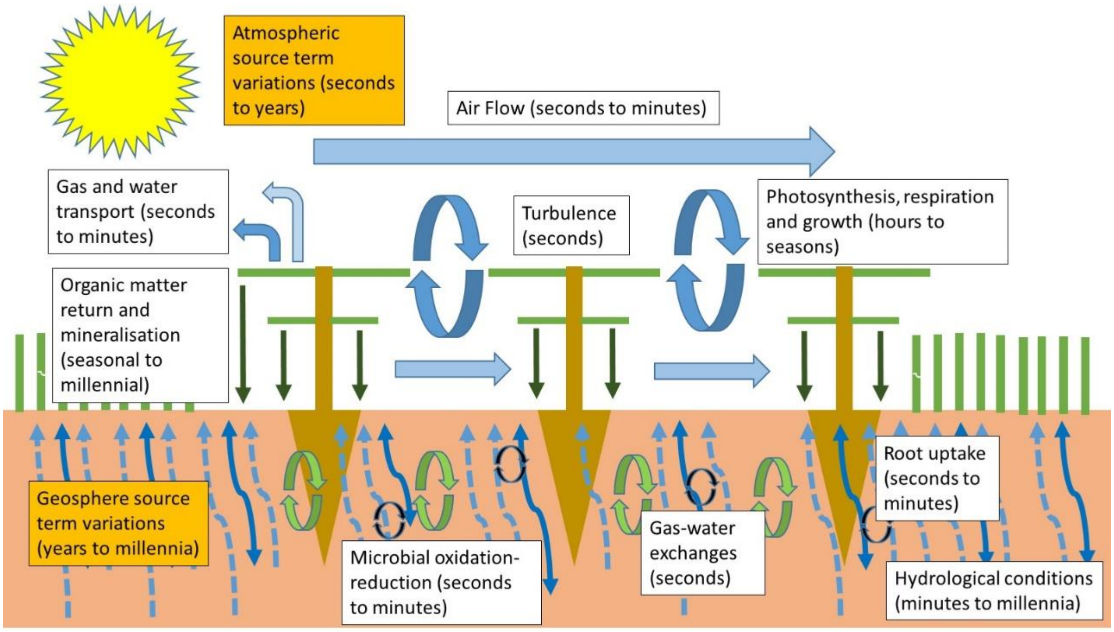

Figure 4. Processes affecting carbon cycling in the biosphere and their associated timescales

The release of C-14 from the geosphere to the biosphere and its subsequent behaviour, potentially giving rise to plant uptake is of particular interest in long-term dose assessments, but there are a number of uncertainties around the behaviour, including the potential for conversion of methane to carbon dioxide and subsequent uptake by plants. RWM have commissioned an experimental programme to investigate the behaviour of methane in the soil rooting zone in agricultural systems.

The research programme consisted of a series of four laboratory and three field experiments on methane (enriched with 25% C-13) transport. The experiment in the second year of field experiments involved a spring wheat planted field. A large improvement in the field technique was associated with this experiment as compared with the first year field experiment, involving a 100 fold more sensitive soil probe. In first season, the field experiment was constrained through the use of tubes to prevent lateral gas movement through the soil. The more sensitive detector meant the second season experiment could be unconstrained experiment such that the vertical and lateral movement of methane was allowed. Experiments were conducted on a loamy sand soil.

Methane was introduced to soils at a rate of 1 mg/day or below and at a depth of around 50 cm and samples were then taken throughout the soil profile at various times following injection. Head space measurements were also taken at a number of times after injection to estimate fluxes of methane and carbon dioxide from the soil. Methane can enter soil from above in mineralised soils with such soils being termed consuming soils. Consumption rates vary considerably for different soils, but grasslands are all largely considered to be consuming soils. The RWM operational environmental assessment also considered methane consumption by soils following atmospheric release of methane with subsequent oxidation in soil and plant utilisation.

Prior to methane injection a range of soil properties and soil methane concentrations at a number of depths were evaluated. The profile of carbon dioxide in the soil is important and was measured, as was the organic matter content in the soil profile, from which the mineralisation rate could be determined. Soil water and gas were also measured from which the soil tortuosity could be inferred to derive the in situ diffusion rate. Further studies of radon diffusion in the experimental soils are being undertaken to confirm these tortuosity estimates using a gas that is not subject to metabolic processes.

The methane inflow and outflow from soils was measured using fixed volume head space chambers. Reducing concentrations were observed indicating a consuming soil. The head space chambers were used to measure methane and carbon dioxide emanation from soils at different times following injection. The processes occurring following injection of methane are illustrated in Figure 5.

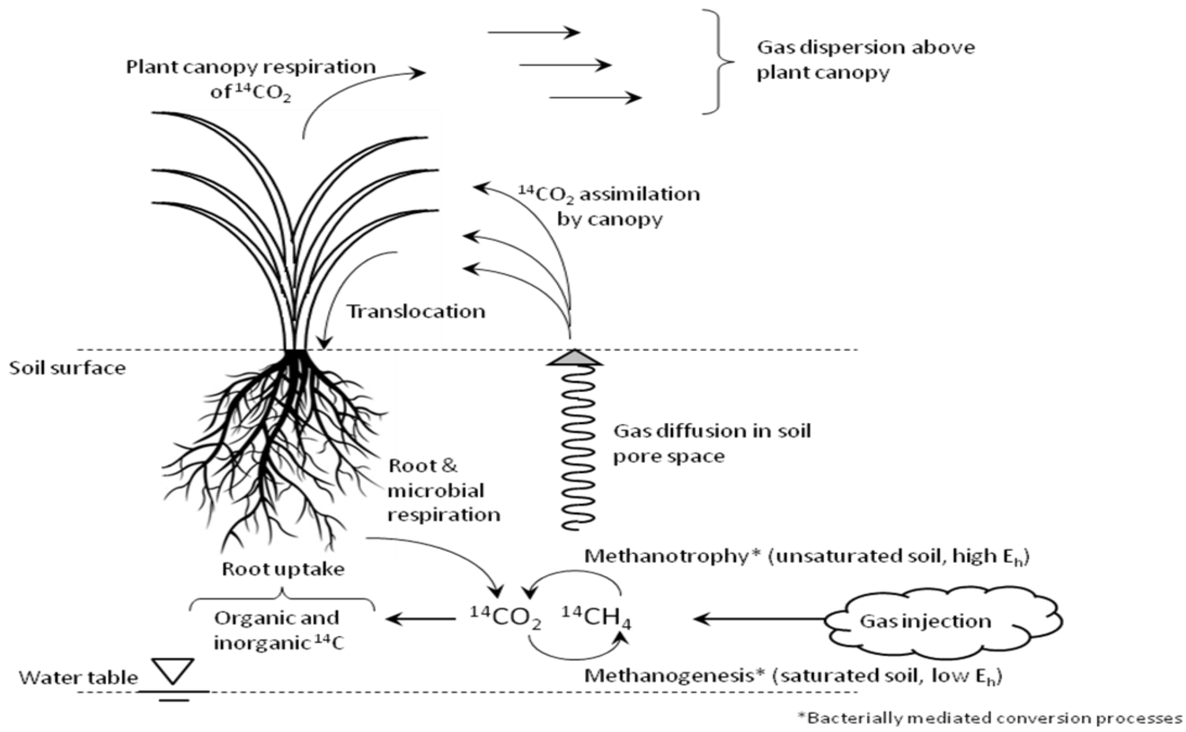

Figure 5. Carbon cycling processes in an agricultural soil following injection of methane.

Perturbation to the soil system associated with sampling of gases from the soil column was been taken into account in the modelling work presented. Only a small volume of methane was injected (less than 1% of soil gas was displaced by injection) so the soil system was not being disrupted as a result of pressure perturbation, but the high concentration within the injected gas (1000 times more methane than is naturally present) allowed the behaviour of the injected methane to be tracked through the soil column.

Illustrative results from wheat field experiments are given in Figure 6. Toward the soil surface, methane concentrations are close to that in air and reduce with depth. The depression is consistent with the flux measured using head space chambers. Upon injection, the methane concentration rose to a peak almost instantly at 40cm soil depth and then at 30, 20 and 10 cm, indicating the applicability of a diffusion model.

In the third laboratory experiment the system was constrained by a cylinder. In contrast to field conditions, a less concave dilution was observed since dilution of methane could only be in one dimension as opposed to three dimensions under field conditions. Methane consumption in soil was observed however such that at 10 cm soil depth, a much lower methane concentration was observed. Results allow the diffusion and consumption rates for methane to be calculated.

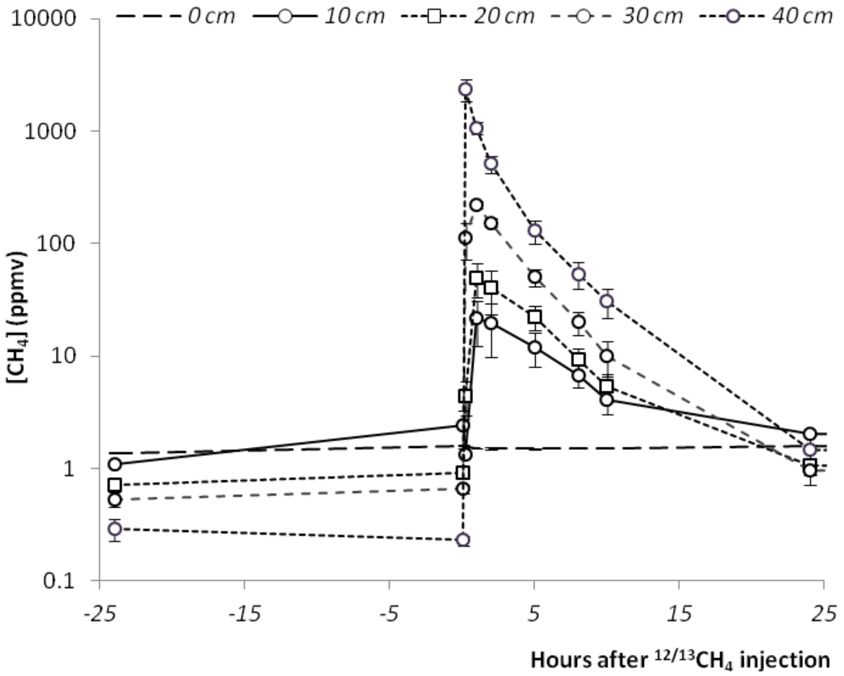

Figure 6. Average bulk methane concentrations measured within gas samples obtained from four depths in vegetated (wheat) field soil following subsoil CH_4 injection. The 0 cm line shows the ambient (free atmosphere) concentration of methane (1.57 ppmv).

Methane concentrations in head space chambers for field experiments are illustrated in Figure 7. The control plots, both vegetated and un-vegetated, show little difference. However, where methane is injected into soils with vegetation and without, there is a greater flux of methane into the head space chamber in the un-vegetated plot. The presence of vegetation has, if anything, slightly decreased methane transport through the system. This will be due to the different soil structure as a result of the presence of vegetation. For example, small channels may be present in soils that are not affected by roots. The influence of roots on soil structure and gas transport will be of interest in a number of situations, such as tree roots in forests and aerenchyma in wetlands.

For the same experiment conducted in the laboratory there was less distinction observed between the vegetated and un-vegetated soils. This may result from sub-lateral transport as well as sub-horizontal differences between field and laboratory experiments; textural change can have greater consequences in a laterally constrained system.

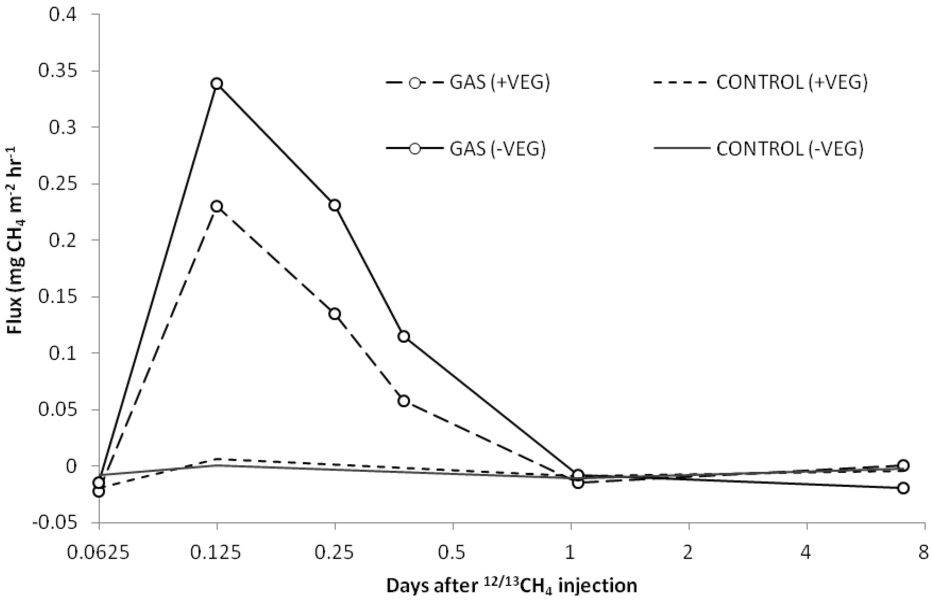

Figure 7. Bulk CH_4 flux from the soil surface into headspace chambers following first subsoil CH_4 injection in vegetated (wheat) field soil.

When methane is first injected into a soil, a high anomaly of δ $^{13}CH_4$ is observed at 40cm that then propagates and attenuates upward through the soil as microbes assimilate the $^{13}CH_4$. In the soil column from laboratory experiment 3, a rapid anomaly was observed at 40 cm that persisted as long as methane persisted throughout the soil. An eventual decline was observed at point of injection. A slower anomaly was observed at higher measurement points in the soil, but a more rapid drop off was recorded as the injected methane exchanges and mixes with methane sourced from the air (Figure 8).

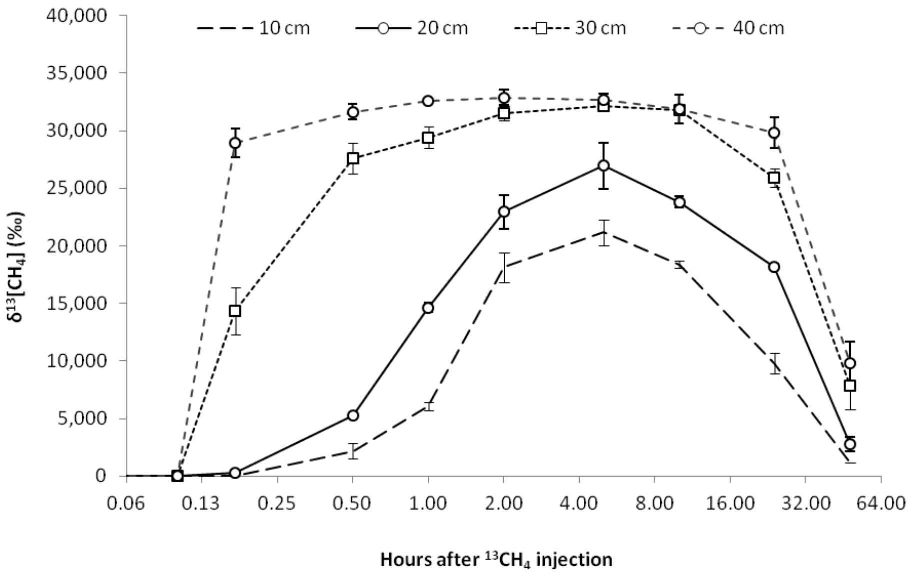

Figure 8. Average δ $^{13}CH_4$ measured within gas samples obtained from four depths in undisturbed, vegetated (ryegrass) soil columns following subsoil CH_4 injection (Laboratory Experiment 3).

In terms of carbon dioxide, at the start of the wheat field experiment, the δ $^{13}CO_2$ was consistent with normal soil (Figure 9). An hour into the experiment a small signal started to appear. Such a small signal was not unexpected. A greater signal was observed in laboratory soil column experiments where no lateral loss could occur (Figure 10).

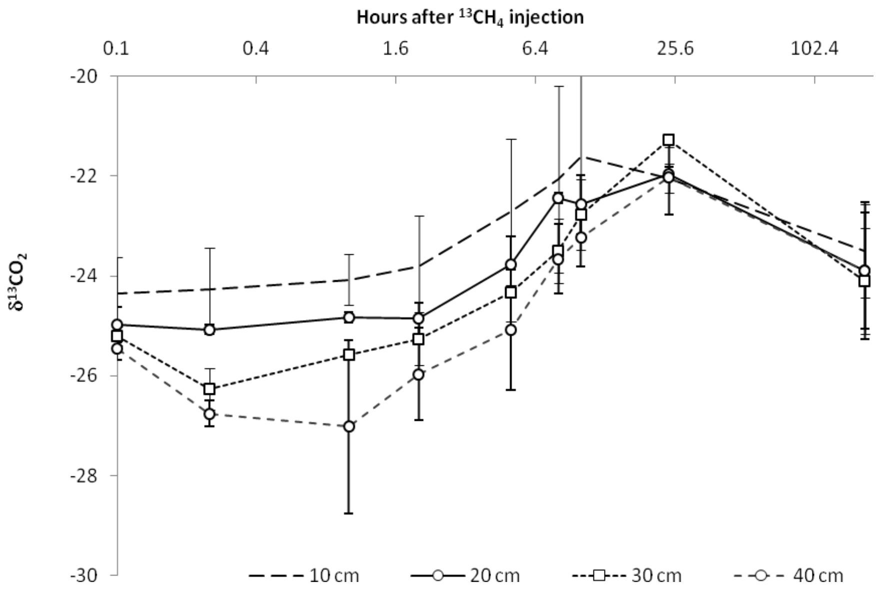

Figure 9. δ $^{13}CO_2$ measured within gas samples obtained at four depths in vegetated (wheat) field soil following subsoil CH_4 injection.

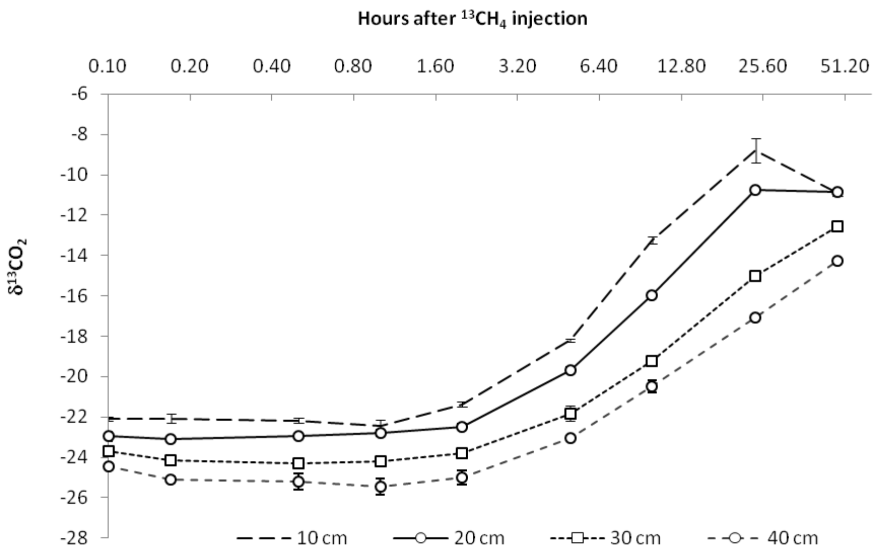

Figure 10. Average δ $^{13}CO_2$ measured within gas samples obtained at four depths in undisturbed, vegetated (ryegrass) soil columns following subsoil CH_4 injection.

A laboratory experiment using $^{14}CO_2$ was also undertaken and a comparison made between the normalised total C-14 and stable $^{12/13}CH_4$ efflux in headspace chambers for the corresponding enriched ^{13}C injection experiment. Both forms were found to increase similarly over time, but the efflux of C-14 continued over a longer duration. This prolonged efflux of C-14 was attributed to it being the combined efflux of $^{14}CO_2$ and $^{14}CH_4$, with only methane being measured in the stable form.

There was a meteorological data station in the experimental field and supporting information on the meteorological conditions during the experimental period are therefore available. A depth profile of soil tortuosity is also available. No C-13 uptake by plants was observed during the experiments. As the experiments were pulse inputs with the isotopic anomaly only persisting for a few days, this was not sufficient to lead to an anomaly in the plants during the growing season.

Overall, results from the laboratory and field experiments were consistent and provide evidence of a rapid diffusion of methane through the soil to atmosphere. The presence of vegetation was shown to affect the soil diffusion regime and there was evidence of the oxidation of methane to carbon dioxide. From the C-14 laboratory experiment it was concluded that $^{14}CH_4$ and $^{12/13}CH_4$ behaviour in terms of soil diffusion are very similar and conclusions drawn from experiments using $^{12/13}CH_4$ can therefore be applied equally when considering the behaviour of $^{14}CH_4$ in soils.

Under agricultural conditions it was concluded that there could be close to 100% conversion of methane to carbon dioxide in soils. The estimated depletion length over which conversion occurs is between 10 and 20cm in unsaturated conditions.

2.6 RECENT C-14 IN THE BIOSPHERE PUBLICATIONS (2012-2015)

Laura Limer presented.

In recent years, several organisations in the nuclear sector have undertaken studies in the area of C-14 dynamics and uptake in the biosphere, including IRSN, Nagra, RWM and SSM. Work has also been undertaken within the BIOPROTA forum. In addition, further academic research has been undertaken outwith BIOPROTA member organisations and related institutes.

Some examples were presented of the work that has been undertaken by waste management organisations and regulators. A BIOPROTA 2012 report on C-14 long-term dose assessment: data review, scenario development and model comparison was published in the SSM reports series (SSM Report 2012-47). The work undertaken within BIOPROTA in relation to C-14 has also been presented at various conferences with associated peer reviewed publications.

IRSN has undertaken a field monitoring campaign around the Cape de la Hague reprocessing plant in order to estimate fluxes of C-14 in a grassland ecosystem (air, rain, plant, soil water) in order to develop a better understanding of the underlying processes in support of the development of the TOCATTA (-χ) model. Various publications have resulted from this work, including:

- Aulagnier et al. (2012). Modelling the transfer of ^{14}C from the atmosphere to grass: A case study in a grass field near AREVA-NC La Hague. Journal of Environmental Radioactivity, 112: 52-59.

- Aulagnier et al. (2013). The TOCATTA-χ model for assessing ^{14}C transfers to grass: an evaluation for atmospheric operational releases from nuclear facilities. Journal of Environmental Radioactivity, 120: 81-93.

- Le Dizès et al. (2012). TOCATTA: a dynamic transfer model of ^{14}C from the atmosphere to soil-plant systems. Journal of Environmental Radioactivity, 105: 48-59.

Nagra has recently undertaken development of their biosphere modelling capability and have updated their conceptual and mathematical model for evaluating the transfer and accumulation of C-14 released to a local aquifer-soil-crop-atmosphere system. The model assumes that C-14 is distributed in the same way as stable carbon and is hence based on masses and fluxes of stable carbon between various carbon pools. The model is described in the 2013 Nagra report NAB 12-26[i].

As presented by Mike Thorne, RWM has undertaken an experimental programme that includes field experiments to study the behaviour of methane in the soil rooting zone in an agricultural system. The programme has resulted in a number of publications, including:

- Atkinson B, Meredith W, Snape C, Steven M and Shaw G (2011). Experimental and Modelling Studies of Carbon-14 Behaviour in the Biosphere: Diffusion and oxidation of isotopically labelled methane ($^{13}CH_4$) in laboratory soil column experiments. NDA Radioactive Waste Management Directorate, report no. AMEC/004041/004, Issue 1.

- Atkinson B, Shaw G, Meredith W, Snape C, Steven M and Hoch A (2012a). Uptake of Carbon-14 in the Biosphere: Data from First Field Experiment, and Modelling of Initial Laboratory Experiments (Progress Report for Year 2). NDA Radioactive Waste Management Directorate, report no. Serco/Nott/e.4041/002 Issue 1.

- Atkinson B S, Meredith W, Snape C, Steven M, Hoch A R, Lever D and Shaw G (2012b). Migration and fate of $^{14}CH_4$ in subsoil: tracer experiments to inform model development. Mineralogical Magazine, 76 (8), 3345-3354.

- Atkinson B S, Meredith W, Snape C and Shaw G (2014). Uptake of Carbon-14 in the Biosphere: Field and laboratory experiments to determine the fate and behaviour of $^{14}CH_4$ injected into agricultural subsoil. Progress Report for Year 3. AMEC/Nott/004041/005, June 2014.

- Hoch A R (2014). Uptake of Gaseous Carbon-14 in the Biosphere: Development of an Assessment Model. AMEC/004041/007, Issue 2.

- Hoch A R, Atkinson B and Shaw G (2014). Uptake of Gaseous Carbon-14 in the Biosphere: Modelling of Field and Laboratory Experiments. AMEC/004041/006, Draft D.

- Hoch A R, Lever D A and Shaw G (2014). Uptake of Carbon-14 in the Biosphere: Summary Report. AMEC/004041/008 Issue 2.

- Shaw G and Atkinson B (2014). A Review of Literature in Support of Experimental and Modelling Studies of Carbon-14 Behaviour in the Biosphere. AMEC/Nott/004041/001 Issue 1.

- Shaw G, Atkinson B, Meredith W Snape C, Steven M, Hoch A and Lever D (2014). Quantifying $^{12/13}CH_4$ migration and fate following sub-surface release to an agricultural soil. Journal of Environmental Radioactivity, 133, 18-23.

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[i] See http://www.nagra.ch/en/cat/publikationen/arbeitsberichte-nabs/nabs-2012/downloadcentre.htm

- Webb S W (2014). T2Plants Version 1.1: Modification of TOUGH2 for near-surface transport of gases, including atmospheric boundary conditions and the effects of plants. AMEC/004041/003, Issue 2.

SSM are also actively considering C-14 modelling for plants in both short and long term release scenarios and are considering whether one model can cover both atmospheric and geosphere source terms. A bilateral collaboration with IRSN allowed model comparison using the Cape de la Hague C-14 data set. A publication detailing the results of the intercomparison has recently been published in the Journal of Environmental Radioactivity. This, and additional publications relating to work undertaken by SSM include:

- Limer, L, Klos, R, Shaw G and Walke R (2013a). Terrestrial biosphere modelling of ^{14}C research, Final report. A Limer Scientific Consulting Limited report, published by the Swedish Radiation Safety Authority, 2013:20.

- Limer L, Klos R, Walke R, Shaw G, Nordén M and Xu S (2013b). Soil-plant-atmosphere modelling in the context of releases of ^{14}C as a consequence of nuclear activities. Radiocarbon, 55 (2-3), 804-813.

- Limer L, Le Dizès-Maurel S, Klos R, Maro D and Nordén M (2014). Improvements in soil-plant-atmosphere modelling of ^{14}C dynamics and the application of two models to data from a nuclear fuel reprocessing plant. International Conference on Radioecology and Environmental Radioactivity (ICRER), 7-12 September 2014, Barcelona, Spain.

- Limer, L M C, Le Dizès-Maurel S, Klos R, Maro D and Nordén M (2015). Impacts of ^{14}C discharges from a nuclear fuel reprocessing plant on surrounding vegetation: Comparison between grass field measurements and TOCATTA-χ and SSPAM^{14}C model computations. Journal of Environmental Radioactivity, 147, 115-124.

Examples of peer reviewed literature from research groups outwith the BIOPROTA community were also presented. Examples include:

- Eyrolle-Boyer et al. (2015). Tritium and ^{14}C background levels in pristine aquatic systems and their potential sources of variability. Journal of Environmental Radioactivity 139: 24-32.

- Melintescu et al. (2013). Carbon-14 transfer into potato plants following a short exposure to an atmospheric $^{14}CO_2$ emission: observations and model predictions. Journal of Environmental Radioactivity 115: 183-191.

- Nakanishi et al. (2014). Seasonal and snowmelt-driven changes in the water-extractable organic carbon dynamics in a cool-temperate Japanese forest soil, estimated using the bomb-^{14}C tracer. Journal of Environmental Radioactivity 128: 27-32.

- Ota M, Nagai H and Koarashi J (2012). A land surface ^{14}C transfer model and numerical experiments on belowground ^{14}C accumulation and its impact on vegetation ^{14}C level. Journal of Environmental Radioactivity 107: 13-22.

- Shaw et al. (2014). Quantifying $^{12/13}CH_4$ migration and fate following sub-surface release to an agricultural soil. Journal of Environmental Radioactivity 123: 18-23.

- Šturm et al. (2012). Carbon isotopic composition ($\delta^{13}C$ and ^{14}C activity) of plant samples in the vicinity of the Slovene nuclear power plant. Journal of Environmental Radioactivity 110: 24-29.

- Tani et al. (2011). Development of a dynamic transfer model of ^{14}C from the atmosphere to rice plants. Journal of Environmental Radioactivity 102(4): 340-347.
- Yankovich et al. (2013). Do site-specific radiocarbon measurements reflect localized distributions of ^{14}C in biota inhabiting a wetland with point contamination sources? Journal of Environmental Radioactivity 126: 352-366.
- Yankovich et al. (2014). Spatial analysis of Carbon-14 dynamics in a wetland ecosystem (Duke Swamp, Chalk River Laboratories, Canada). Journal of Environmental Radioactivity 137: 173-180.
- Naofumi et al. (2013). Radiocarbon Concentrations in Environmental Samples Collected Near the Spent Nuclear Fuel Reprocessing Plant at Rokkasho, Aomori, Japan, During Test Operation Using Spent Nuclear Fuel. Health Physics 105(3): 236-244.
- Tani et al. (2013). A Dynamic Transfer Model for the Estimation of ^{14}C Radioactivity in Japanese Radish (Daikon) Plants. Health Physics 105(2): 121-127.
- Yoshihito et al. (2013). Daily Radionuclide Ingestion and Internal Radiation Doses in Aomori Prefecture, Japan. Health Physics 105(4): 340-350.
- Gillon et al., (2012). Controls on ^{13}C and ^{14}C variability in soil CO_2. Geoderma 189-190: 431-441.
- Tatzber et al. (2012). ^{14}C-labeled organic amendments: Characterization in different particle size fractions and humic acids in a long-term field experiment. Geoderma 177-178: 39-48.
- Hartley et al. (2013). The age of CO_2 released from soils in contrasting ecosystems during the arctic winter. Applied Soil Ecology 63: 1-4.
- Olsrud and Christensen (2011). Carbon partitioning in a wet and a semiwet subarctic mire ecosystem based on in situ ^{14}C pulse-labelling. Applied Soil Ecology 43(2): 231-239.
- Tian et al. (2015). Aggregate size and their disruption affect ^{14}C-labeled glucose mineralization and priming effect. Applied Soil Ecology 90: 1-10.
- Tipping et al. (2012). $N^{14}C$: A plant–soil nitrogen and carbon cycling model to simulate terrestrial ecosystem responses to atmospheric nitrogen deposition. Ecological Modelling 247: 11-26.
- Tefs and Gleixner (2012). Importance of root derived carbon for soil organic matter storage in a temperate old-growth beech forest – Evidence from C, N and ^{14}C content. Forest Ecology and Management 263(1): 131-137.
- Rzeznik-Orignac and Fichet (2012). Experimental estimation of assimilation rates of meiofauna feeding on ^{14}C-labelled benthic diatoms. Journal of Experimental Marine Biology and Ecology 432-433: 179-185.
- Chen et al. (2014). Fate of ^{14}C-labeled dissolved organic matter in paddy and upland soils in responding to moisture. Science of the Total Environment 488-489: 268-274.

Discussion

It was noted that the Food Standards Agency in the UK, which has a remit to develop an understanding of contaminants in human health chains, requires models to evaluate potential impacts on people should pollution incidents occur. Currently they use two different approaches to evaluate C-14, depending on whether a spike release has occurred or whether the release relates to a long-term discharge. For the latter, an equilibrium approach is applied. For short-term releases, the FSA has invested in the PRISM tool for which C-14 modelling capability was added in 2005-6. PRISM includes soil hydrology and has specific consideration of roots and other parts of plants. Some data may be available from the FSA to support model comparison activities.

2.7 EXPERIMENTAL DATA SETS FROM A GRASSLAND ECOSYSTEM IN THE VICINITY OF LA HAGUE REPROCESSING PLANT: MAKING RADIOECOLOGICAL DATA AVAILABLE

Severing le Dizes-Maurel presented.

The VATO (validation of TOCATTO model) project consisted of two programmes; development of the TOCATTA model for a terrestrial ecosystem, and experimental campaigns to look at the transfer to plants of tritium and C-14, released to atmosphere from the la Hague reprocessing plant, accounting for air concentrations (both day and night), weather conditions and land use. The aim was to better understand the underlying processes involved and to identify any gaps in the TOCATTA model.

TOCATTA is a dynamic transfer model for C-14 in air-soil-plant systems that runs on a daily time step. The model employs equilibrium assumptions for plant-air concentrations and two soil types (sand and clay) are considered. The modelling platform can be used for different crop types. The model is primarily driven by $^{14}CO_2$ concentrations in air and daily meteorological data. Organic C-14, mostly emitted as methane plays no direct role in photosynthesis and is not considered in the model.

The experimental campaign focussed on a ryegrass plot located 2km downwind (in the predominant wind direction) of the la Hague site, which releases both tritium and C-14. Releases from the facility induce greater C-14 concentrations in plants as compared with the natural background for the area. A laboratory was installed at the site and measurements taken from October 2006 to July 2008. A 10m mast is installed at the site that measures wind direction and speed. There is also a weather station present. The sampling programme included air concentrations at 1.5 m above the soil and monthly monitoring of ryegrass and soil (with ryegrass being cut to around 1 cm to mimic grazing) with parallel monitoring of meteorological data. Transfer of C-14 to cow milk was also monitored, but with only limited sampling. A continuous monitoring device for Kr-85, as a tracer of the plume from the facility, was employed, with C-14 and tritium being then sampled using a bubble trapping device that was analysed monthly. Grass was sampled from the same 1 m^2 plot each month, but grass throughout the field was also cropped such that the canopy within the sampling plot was not affected by growth of the surrounding grass. The data that are available as a result of the monitoring programme, that can be made available to the assessment community, include a range of atmospheric data (presented in Table 1) and information relating to the source term, plant biomass and C-14 activity measured in different media at the site (summarised in Table 2). Different analysis methods for C-14 in grass were employed in each of the three years of sampling.

Table 1. Atmospheric data and monitoring frequency.

Data type	Variables	Units	Time period	Frequency
Meteorological	Temperature	°C	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins 30 mins [b]
	Humidity	%	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins 30 mins [b]
	Global radiation	J cm^{-2}	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins 30 mins [b]
	Wind speed	m s^{-1}	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins [b] 30 mins [b]
	Wind direction	deg	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins [b] 30 mins [b]
	Air pressure	Hpa	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins [b] 30 mins [b]
	Precipitation	mm	28/6/06 – 27/12/06 11/1/06 – 18/12/08 [a]	30 mins 30 mins [b]
	Past wind (Vent passé)	km	11/1/06 – 18/12/08 [a]	30 mins [b]
	Average 30 min wind speed	m s^{-1}	11/1/06 – 18/12/08 [a]	30 mins [b]

Table 2. Summary of available data and monitoring frequency relating to source term, plant biomass and C-14 activity at the monitoring site.

Data type	Variables	Units	Time period	Frequency
Source term	^{14}C releases from three chimneys [c]	Bq	07/1/06 – 31/12/08	Weekly
	^{85}Kr measurements at the field site	Bq	01/10/06 – 01/1/08	Hourly
	^{85}Kr measurements at the field site	Bq	31/1/06 – 21/12/08	Monthly
	^{14}C measurements at the field site	Bq	31/1/06 – 21/12/08	Monthly
Plant biomass	Biomass grass harvested [d]	g m^{-2} fw	01/8/06 – 04/12/08	~ Monthly
	Biomass grass harvested [d]	g m^{-2} dw	01/8/06 – 04/12/08	~ Monthly
^{14}C activity at the site	Specific activity of soil surface (0-3 cm)	Bq kg^{-1} C	01/8/06 – 31/8/07	~ Monthly
	Specific activity of deeper soil (0-20 or 3-20 cm)	Bq kg^{-1} C	29/6/06 – 31/8/07	Jun, Aug, Nov
	Carbon content of the soil	%	29/6/06 – 31/8/07	As ^{14}C meas.
	Grass	Bq kg^{-1} C	01/8/06 – 01/3/07	~ Monthly

Measurements indicate that C-14 concentrations in air and grass fluctuate greatly, but soil concentrations, whilst relatively high (i.e. around 70% larger than typical background values), are fairly constant. The variation in air and grass results from variation in wind direction and reduced reprocessing activities in the summer when staffing is reduced for holidays. During these periods, grass concentrations reduce almost to background whereas soil concentrations remain high. This may result from retention in soils in an organic form with only slow degradation over time to a more bioavailable form. Some degradation must occur or the soil concentration would continue to build up over time as the facility continues operation.

The data set has been used to test and validate the TOCATTA model and can be shared with others for model testing purposes. Indeed, the data has already been shared with SSM and a paper has been submitted on the results of a resultant model-model and model-data intercomparison[j].

Work is currently being undertaken to obtain similar data for tritium and will include organic tritium formation in plants from vapour, rain and soil water. Tritium concentrations in air are being reconstructed in hourly time steps, taking into account wind direction and the different release chimneys of the site. The data set for tritium is being shared with Working Group 7 of the IAEA MODARIA programme with 3 models being applied to the data set.

Discussion

With regard to the reconstruction of C-14 air concentration data from Kr-85 measurements, it was noted that, for the la Hague site, Kr-85 is a good tracer for C-14, with 90% of fuel having a similar burn-up and hence stable release ratio. The ratio may not however be constant for other sites, as C-14 release rates would be expected to vary according to different processes. With regard to tritium, the ratio of inorganic tritium to Kr-85 would be expected to be stable, but not so for organic tritium.

There was interest expressed by workshop participants in the use of the IRSN data set by others outside the initial IRSN-SSM collaboration. Furthermore, the use of alternative datasets, should they be identified and available, would be of interest to IRSN, allowing further model testing using the TOCATTA-χ model.

2.8 SSM/IRSN MODEL INTERPRETATION OF FIELD WORK

Laura Limer presented.

SSM and IRSN have collaborated on a model-model, model-data intercomparison, focussing on the monitoring programme of IRSN (see section 2.7 above).

SSM has been interested in C-14 dose assessment for some time with a review of models being undertaken in 2011, followed by development of a C-14 assessment model, the Swedish soil-plant-atmosphere model for C-14 (SSPAM^{14}C). The model was initially tested against C-14 data arising from studies undertaken by Imperial College, London. Further model testing then began in 2013 with the IRSN la Hague field measurement data. SSM also has interest in C-14 in the aquatic environment and a model testing exercise was performed in 2013 using data from Canadian Shield Lake experiments.

[j] Since the meeting, this paper has been accepted for publication.

For the terrestrial modelling comparison, both SSM and IRSN models were applied independently to the data set and output from each models compared. The principal properties of each of the models are presented in Table 3.

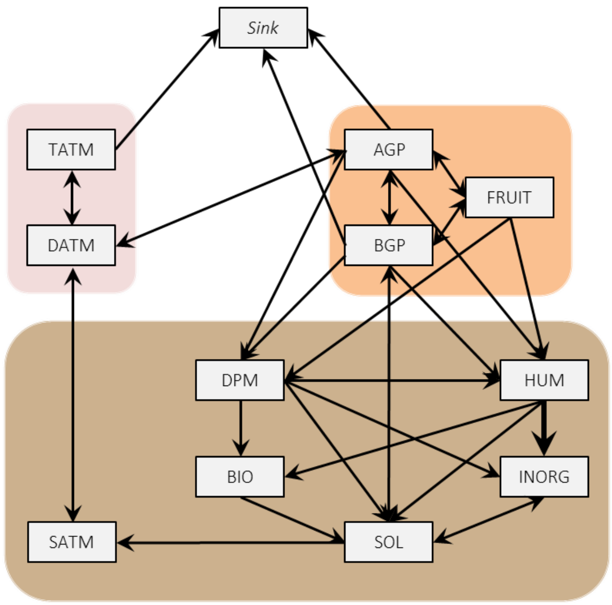
Table 3. Model properties: SSPAM^{14}C and TOCATTA-χ

Model property	SSPAM^{14}C	TOCATTA-χ
Compartments in plant	Aboveground plant Belowground plant Fruit	Substrate (sap) pool (i) Shoot structural dry matter pool (ii) Root structural dry matter pool (iii)
Sub-models	Plant (S1), Soil (S2), Atmosphere (S3)	Plant (S1), Soil (S2)
Implemented processes in (S1)	Biological growth (P1) Photosynthesis (P2) Growth & maintenance respiration (P3) Senescence (P4)	Biological growth (P1) Photosynthesis (P2) Growth & maintenance respiration (P3) Senescence (P4)
Isotopic equilibrium	Between lower atmosphere and aboveground plant	Between air and P2(i) Between (i) and P1(i) Between (i) and P3(i)
Time-step running	24 hours	1 Hour
Input data	Daily ^{14}C in air Relative growth rates between harvests Wind speed	Hourly ^{14}C in air Hourly temperature Hourly net solar radiation Daily sun zenith angle
Influence of specific activity of air	Instant	20 day moving average
References	Limer et al. (2013). SSM Report 2013:20.	Aulagnier et al. (2013). Journal of Environmental Radioactivity 120: 81-93.

The SSM model considers operational discharges to the surface environment (atmospheric release and surface water discharge) and releases from below ground relating to radioactive waste disposal (upwelling groundwater, irrigation and gaseous release from below). The IRSN model is focussed on operational discharges. This difference in focus also drives the difference in time steps. When considering long term discharges, a single plant compartment may be adequate, whereas for operational discharges there may be more interest in multiple plant compartments.

SSPAM^{14}C is implemented in Amber whereas the TOCATTA-χ model is implemented in Symbiose. The original TOCATTA model uses a single plant compartment and daily time step whereas TOCATTA-χ is more process oriented and hence more detailed, using three plant compartments (plant dry matter, shoot structural dry matter and root structural dry matter). The SSM model considers three plant compartments, the above ground plant and the below ground plant and fruiting bodies. The overall structure of the SSM model is presented in Figure 11.

The SSM and IRSN models differ with regard to equilibrium assumptions and time steps. For example, the SSM model assumes an instant influence of air C-14 concentrations on that in plants whereas the IRSN model uses a 20 day moving average to take account of the finite timescale of C-14 accumulation and retention by plants. The SSM model uses annual process data rather than considering seasonality. A 1% total carbon uptake from roots to plants is also assumed. Methane uptake is not considered.

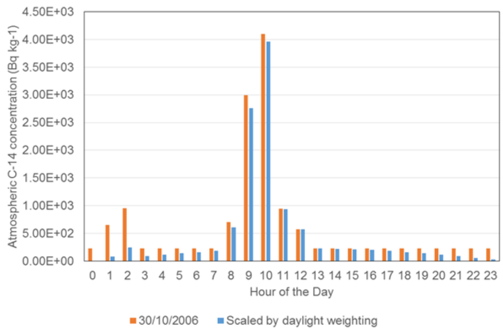


- Soil
  - DPM – decomposable plant material
  - HUM – humic substances
  - BIO - biota
  - INORG – inorganic material
  - SOL – dissolved C
  - SATM – soil atmosphere
- Plant
  - AGP – aboveground plant
  - BGP – belowground plant
  - FRUIT – fruit
- Atmosphere
  - DATM – diffusive atmosphere
  - TATM – turbulent atmosphere
  - *Sink*

Figure 11. Structure of the SSM SSPAM^{14}C model.

Both the SSM and IRSN models were applied to the IRSN La Hague field measurement data with the objective of comparing the output of both models in terms of soil and plant concentrations as derived from air concentration data. Due to the differences in model time steps, hourly data on C-14 in air had to be back-calculated to derive daily time step data for input to the SSM model. A daylight hour-weighted mean C-14 concentration was calculated in order to take account of the C-14 that the plant sees during the photosynthetic period. The approach to calculation is illustrated in Figure 12. Alternative averaging approaches give rise to different atmospheric C-14 concentrations (Figure 12).

$$\mathit{Daily\ Value}(i) = \frac{\sum_{j=0\ldots23} \sin\left(\frac{j}{24}\pi\right)\cdot \mathit{Hourly\ Value}(i,j)}{\sum_{j=0\ldots23} \sin\left(\frac{j}{24}\pi\right)\cdot \mathit{Hourly\ Value\ Available}(i,j)}$$



Average (30/10/2006)	Atmospheric C-14 concentration
Unweighted daily average	6.12E+02
Weighted daily average (with sin(x) scaling on denominator)	7.55E+02
Weighted daily average (without sin(x) scaling on denominator)	4.80E+02

Figure 12. Approach to deriving daylight hour-weighted mean C-14 air concentration (Bq/kg) for the SSM model.

Results illustrate that the TOCATTA-χ model is more accurate in predicting plant concentrations than the non-process based TOCATTA model (Figure 13). A more mechanistic consideration of processes therefore led to improved model predictions.

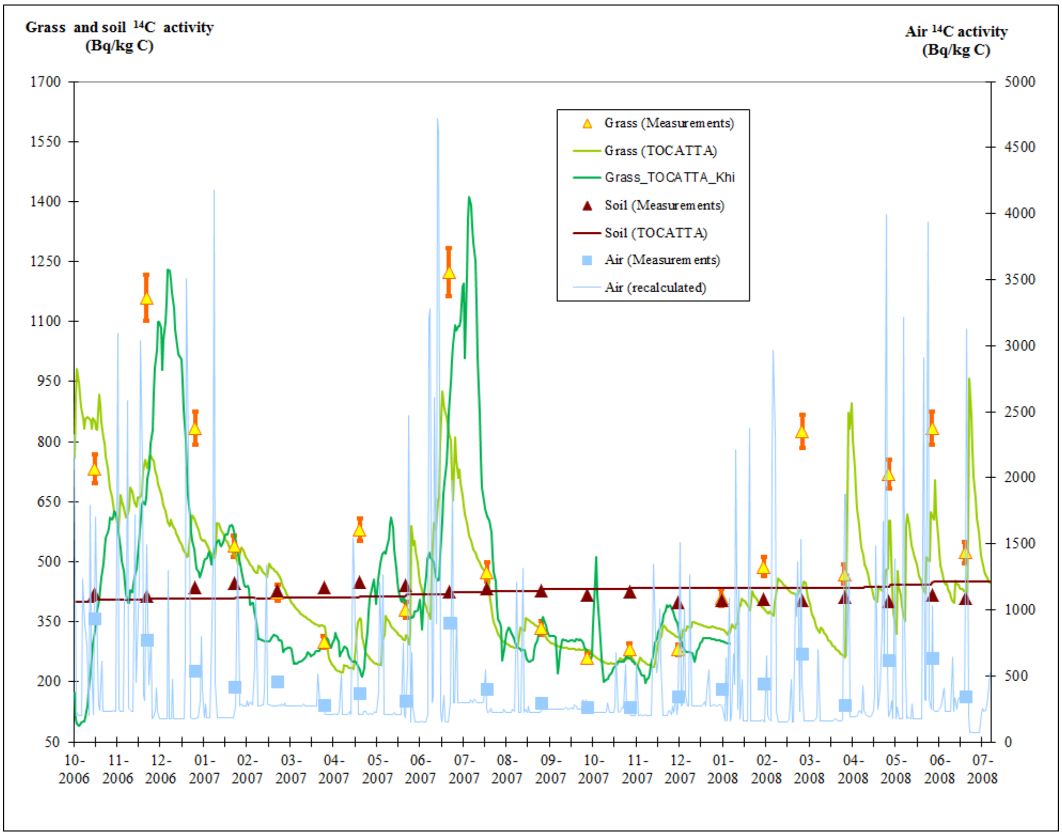


Figure 13. Comparative plant concentration results for the TOCATTA and TOCATTA-χ models.

For the SSM model, two approaches were taken. One was to adopt a steady state assessment approach whereby biomass was fixed and constant fluxes assumed. The other was to take account of seasonal plant dynamics and transfers. Results (Figure 14) indicate that the long-term approach over-predicted plant concentrations. Taking account of seasonal processes (e.g. reduced plant growth in winter months) led to improved predictions. A 20-day moving average of C-14 concentrations in air, combined with seasonal plant dynamics was also considered. This approach further improved model predictions over the long-term approach.

The seasonal dynamics were calculated on the basis of production rates measured at the site once a month in combination with literature data on the ratio of plant respiration to gross photosynthesis and how this varies over the course of a year for pasture.

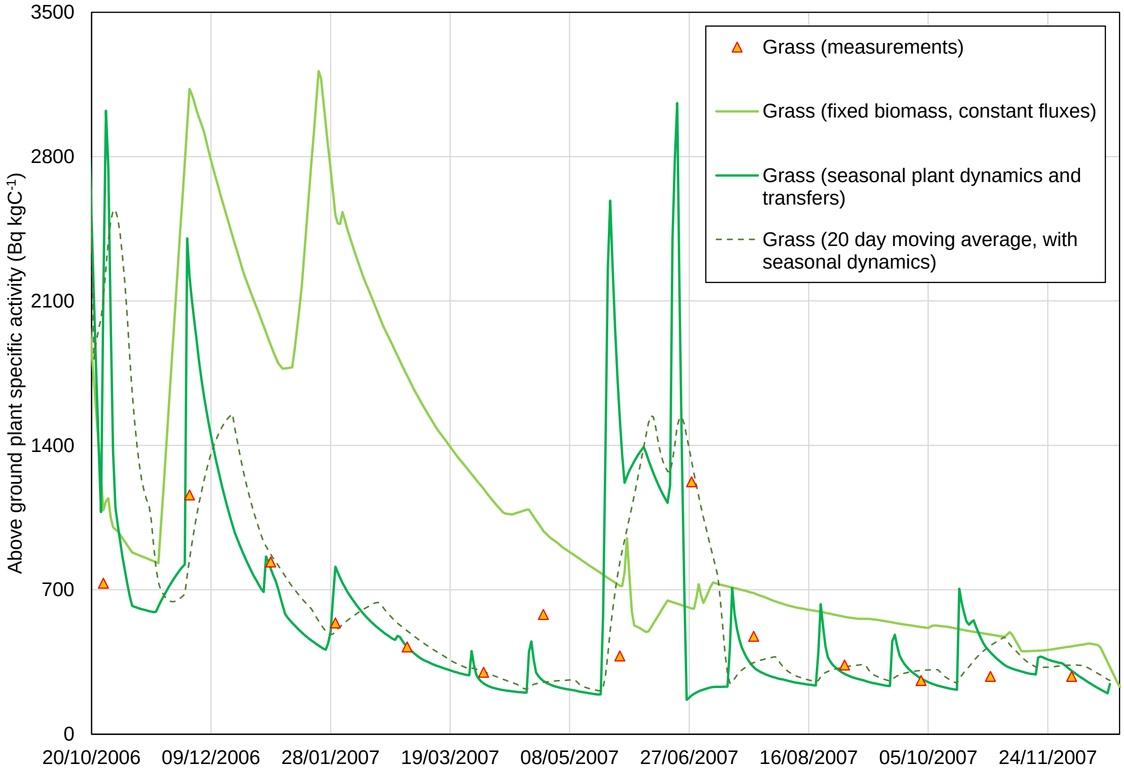


Figure 14. SSM plant concentration predictions using different air averaging and plant dynamics assumptions.

Comparison of both models with field measurements show reasonable agreement when the SSM model is run with the 20 day moving average and seasonal dynamics (Figure 15). A difference in starting concentrations is observed however, that results from differences in atmospheric assumptions. The correlation between model curves was investigated and results (Table 4) indicate that the inclusion of seasonal dynamics in the SSM model resulted in improvements over the use of annual assumptions, but the improvement was not large. The behaviour of the models was more aligned when seasonal dynamics and the 20 day rolling average were considered. Concern was raised however that, by averaging air input data and averaging within the model itself, double counting may occur and this could be investigated. The TOCATTA-χ model plant concentration predictions were always in good agreement with field measurements. There is however a phase shift in air to plant concentrations in the early stages that is not observed in the later stages of the model simulation. It was noted that the IRSN model takes account of meteorological conditions and this may help to explain why the output is more aligned to field measurements. The results suggest that long-term average data points can only be achieved in models if short-term meteorological data are taken into account. It is not feasible however to run such a detailed model for long-term assessments and appropriate mechanisms to simplify approaches must therefore be considered.

The IRSN and SSM models adopt different approaches with regard to the representation of processes. The IRSN model focuses on operational releases and is detailed in its consideration of processes whereas the SSM model has a simplified representation of processes to allow application to long term assessments.

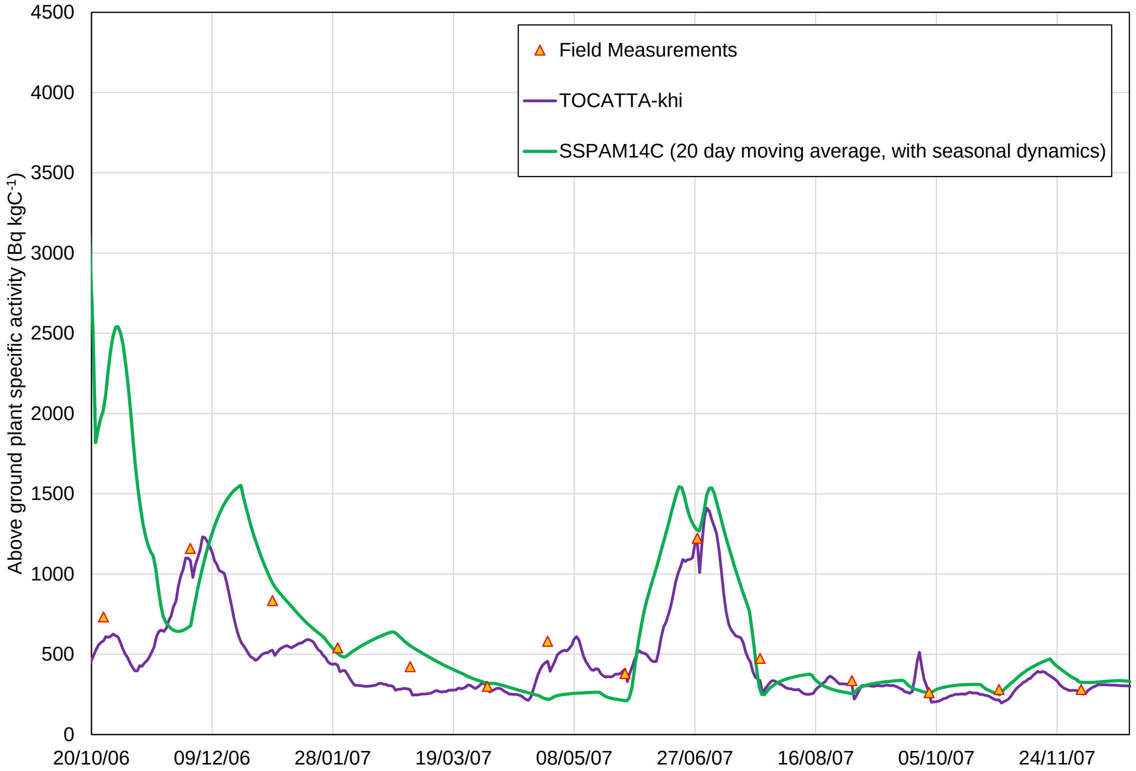


Figure 15. Results of the TOCATTA-χ and SSPAM^{14}C models in terms of plant C-14 concentrations and comparison with field measurements.

The soil activity concentration at the field site was fairly constant throughout the monitoring period. As such, it was not possible to compare the output of the models to determine whether a complex representation of processes in soil is required or whether a more simple approach is adequate.

Table 4. Correlation between the SSM and IRSN models under different averaging assumptions.

MODEL 1	MODEL 2	PEARSON CORRELATION COEFFICIENT	
		Full time series	**Results from 30/11/06 onwards only**
TOCATTA-χ	SSPAM^{14}C (constant biomass and transfers)	0.38	0.32
TOCATTA-χ	SSPAM^{14}C (seasonal dynamics)	0.46	0.68
TOCATTA-χ	SSPAM^{14}C (seasonal + 20 day moving average)	0.45	0.82

Discussion

With regard to assumptions to derive weighted daily air concentrations as input to the SSM model, it was noted that plants respire at night, but no uptake occurs. There is therefore a releasing system during night time. Whether or not this should be specifically included in models should be considered. Furthermore, plant photosynthesis over time is more complex than assuming a sinusoidal variation over

24 hours or a constant rate over 12 hours of daylight. The photosynthetic process begins around dawn and increases until optimum daylight conditions are achieved. The photosynthetic process then reduces as light conditions become sub-optimal once more. In some assessment cases, it may be appropriate to take into account this variation.

2.9 ANALYSIS OF C-14 TRANSPORT FROM A CONTAMINATED GROUNDWATER SOURCE

Jon Napier presented.

A research programme has been undertaken at the AECL Chalk River site in Canada as part of a PhD programme. The objectives were to collect data on carbon, both stable and C-14 forms, in groundwater, soil gas and plants and from this, to create an independent data set specific to C-14. The initial focus was on inorganic forms of carbon, but organic forms were added as the programme progressed. Waste Management Area C (WMA-C) was the focus, which is upstream in terms of groundwater of Duke Swamp. It was the upstream area between the waste disposal area and the swamp that was studied rather than the swamp itself.

The WMA-C area is comprised of a trench within which low level waste has been disposed of in shipping containers. Leakage occurred from these containers almost as soon as they were disposed of resulting in the release of both radioactive and non-radioactive contaminants. High concentrations of both tritium and C-14 have been observed at the site. Most of the tritium contamination has been transported in groundwater to Duke Swamp, although some degassing may have occurred.

There are two further contaminant sources to northwest of WMA-C, a thorium pit that received waste from 1955 to 1960 and a nitrate plant that had an accidental discharge of waste directly to an infiltration pit. It is estimated that some 60 TBq of Sr-90 was released as a result of this accident. The different plumes, relative to Duke Swamp, resulting from these sources are illustrated in Figure 16.

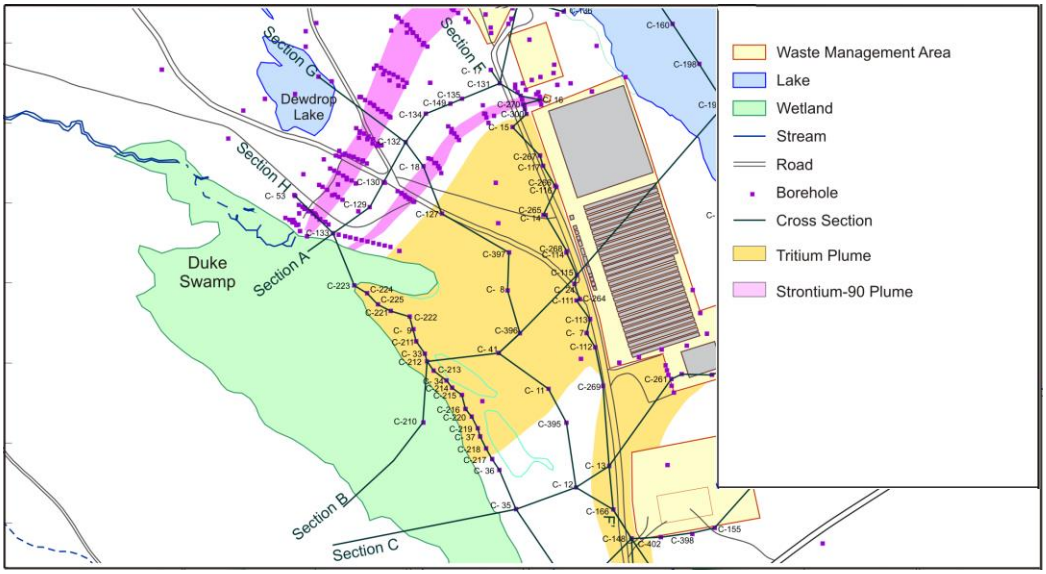


Figure 16. Map detailing contaminant plumes from WMA-C and additional contaminant sources.

The site is characterised by a sandy soil overlaying glacial till that was deposited during the last ice age. Groundwater is sourced from rainwater (ca. 800 mm/y) and is also fed from Lake 233. The movement of groundwater is around 150 m per year.

A two year sampling programme has been undertaken with a variety of parameters collected, as indicated in Table 5. Samples were located in areas expected to be contaminated and at test wells, according to their location relative to expected plumes.

Table 5. Parameters sampled in the Duke Swamp study.

Variable Name	Units
Plant C-14 Activity	^{14}C Bq g^{-1} C
Soil Gas Sampling Depth	m
Soil Gas Carbon Dioxide	ppm
Soil Gas Combustible Gas	ppm
Depth to Groundwater	m
Groundwater C-14 Activity	^{14}C Bq L^{-1}
Soil Gas C-14 Activity	^{14}C Bq L^{-1}
Dissolved Organic Carbon	mg L^{-1}
Dissolved Inorganic Carbon	mg L^{-1}
Plant Dry Mass	g
Plant Wet Mass	g

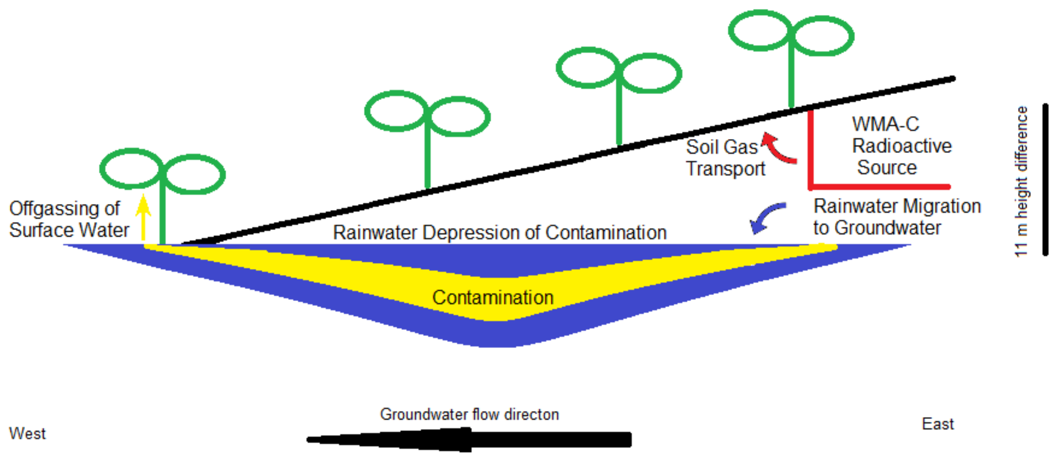
Some differences in sampling and analysis occurred between the two sampling years. For example, the procedure for soil gas collection and analysis was improved with the efficiency being 99% in the second year of sampling. Multi-level soil gas samplers were also installed for the second year and the carbon dioxide detector location was altered. Soil gas samplers were placed to ensure that gas depletion of one area did not affect another. Carbon was isolated from all samples; other contaminants were not monitored.

A diffusion gradient was observed for one sampling site. It had been assumed that C-14 would be near the groundwater surface, but was not the case. Rather, C-14 was measured in soil gas near groundwater, but not in groundwater itself.

Plant (bracken fern) activity concentrations were corrected for each sampling point in order to calculate the excess carbon activity over background. Correction data were derived from provincial background measurements. After correcting there was a notable void of excess C-14 in the upper plume. C-14 activity was observed close to the swamp and near the WMA-C site, but there was a void between these that had not been expected. The two highest data points were recorded close to the swamp, which had been expected. Following this observation, measurements were subsequently made along four transects to derive a horizontal view of off-gassing. Results are illustrated in Figure 17.

From the monitoring results it appears that continued rainfall depresses the C-14 contamination plume, leading to a reduced soil degassing away from the WMA-C until the plume reaches the swamp area (Figure 17a). Plant activity concentrations are therefore greatest toward the swamp.

a)



b)

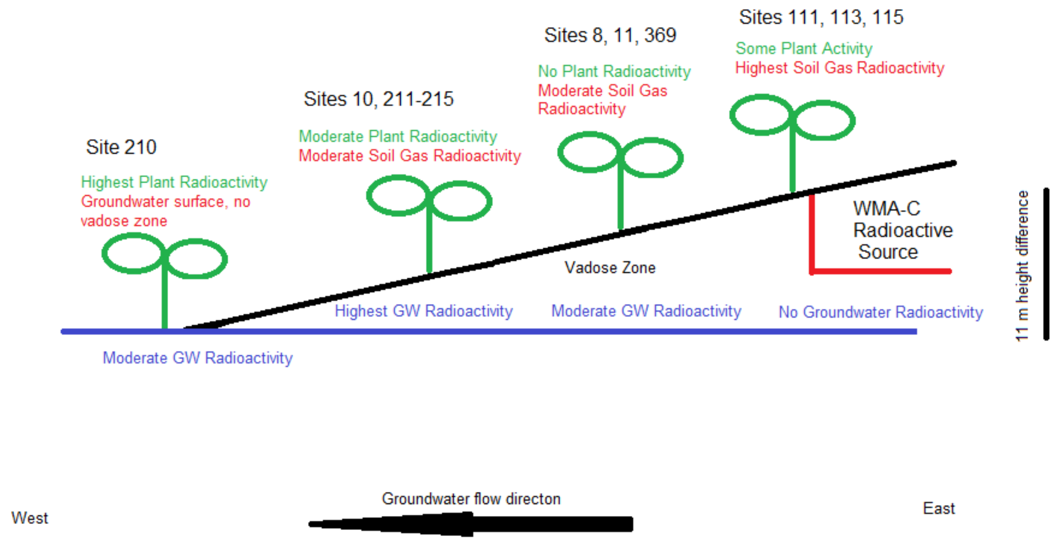


Figure 17. C-14 behaviour in the Duke Swamp upland area (a) and the conceptual view (b).

With monitoring and analysis approaches varying between the two years of sampling, an efficiency correction was required for those samples taken in the first year. Combustible gas concentrations were not recorded in the first year and so it was assumed that the data were consistent with those in the second year. Water activity concentrations from 2014 were also used to represent those at the same sampling sites in 2013. Year to year correlations have been calculated and most parameters were highly correlated, with the exception of DIC in groundwater. Correlations were observed between plant C-14 concentrations and those in groundwater and soil gas with groundwater depth.

A best fit statistical model for the entire data set was chosen using a statistical analysis package. There were some significant variables identified, including dissolved organic carbon (DOC) and sample depth. If the swamp site was excluded, more variables were identified. By removing zero data points from sites not within the plume zones, overall data points were reduced by around 50% which led to an improved best fit model that indicated that the key parameters governing plant C-14 activity were groundwater depth and soil gas activity concentrations. Further analysis considered whether the zero data were located east or west of the active areas. For the east area data, i.e. leaving WMA-C, plant C-14 concentrations were most strongly correlated with soil gas activity. Moving toward the swamp area, the best indicator for plant C-14 activity concentrations was water depth. The results support the hypothesis that groundwater contaminated from the WMA-C is overlain by rainwater in the forest area that emerges toward the swamp.

Distance as an indicator was subsequently considered. The distance of each sampling point from WMA-C and from each point to the edge of the swamp was calculated. When these distances were incorporated within the statistical model, predictors for plant C-14 concentrations changed, with distance being an important predictor.

Discussion

Questions were raised as to how distance should be included in the model where more than one contaminant plume is present in an area. The statistical modelling approach presented differs from that routinely applied in long term dose assessments where the focus is largely on different model compartments and the interactions between these. It was however noted that one of the variables being investigated was soil gas (Bq/l) which was used to represent C-14 transport in the model. If distance is also added to the model then the addition is effectively a correlate of the soil gas variable. It is possible that soil gas may not be linearly related to distance and, as such, more complexity may be being added which reduces the correlation. Consideration should be given to the actual process by which C-14 is moving down the slope. For example, groundwater C-14 is likely to be correlated with distance, but movement may be deflected laterally which would reduce the correlation with distance and slope. It was therefore suggested that distance may not be an appropriate variable to include in an explanatory statistical model. To further model the hydrology and gas transport in the system, the monitoring of tritium and organically-bound tritium in groundwater and gaseous release would be beneficial.

With plant C-14 concentrations being correlated with soil gas C-14, consideration could be given to the height of the plant canopy along the sampling transect and whether this affects plant C-14 concentrations. A further factor governing C-14 behaviour could be the soil redox conditions. Variability in redox along the transect could be driving the degassing of C-14 from soils. There could also be different forms of C-14 present that could explain some of the variation observed. For example, more volatile forms may be present in the vicinity of WMA-C leading to an initial release of C-14 in this area with a second pathway occurring toward the swamp from an alternative C-14 form.

Finally, it was noted that the upflow data presented could be combined with wetland data for the swamp from research undertaken by Tamara Yankovich, mentioned in Section 2.6 to give a more complete data set that may be of interest for model testing by the assessment community.

2.10 PROGRESS ON A VOXEL PHANTOM MODEL OF A PINE TREE

Caitlin Condon presented.

Interest in environmental protection from ionising radiation has increased in recent years and, in 2008, ICRP publication 108 was released that focusses on biota and the environment. ICRP 108 highlights the need to link exposure to dose and dose to effect for a set of Reference Animals and Plants (RAPs). These RAPs were selected to represent the major types of animal and plant in a generalised way that can be used to cover a variety of species. The effects endpoints of interest are mortality, morbidity and reproductive success. In total, 12 RAPs were selected:

- Deer
- Rat
- Duck
- Frog
- Trout
- Flatfish
- Bee
- Crab
- Earthworm
- Pine Tree
- Wild Grass
- Brown Seaweed

Currently, doses are evaluated on the basis of simplified geometric representations of each of the RAPs. In order to improve the evaluation of dose, an MSc project has begun, focussing on the pine tree with the objective of creating a voxel phantom representation of this RAP.

As a result of the Chernobyl accident, a lot of data on pine tree death relative to radiation exposure has been gained. Studies in the area have shown that the LD_{50} for pine tree could be as low as 4.6 Gy, which is similar to that for people. There is however a wide range of LD_{50} data depending upon factors such as age. The large range in data prevents clear conclusions from being drawn. A Sr-90 dose of between 20 and 40 Gy was observed to be lethal for all pine trees.

Two further sites are known to present radiation exposure to pine trees. Savannah River Site (SRS), located in the United States, hosted five nuclear reactors plus chemical separation facilities. Historical contamination released from these facilities to the immediate surrounding environment has the potential for forests on the SRS to be exposed to radionuclides. Forests around Fukushima in Japan have been exposed to radiation as a result of the Fukushima Dai-ichi reactor accident that led to an aerial plume of released radionuclides.

Pine trees are the pillar of many ecosystems and define the ecosystem they are in. Pine trees reach maturity at between three and five years and are very long lived. They are an important component of ecosystems, providing shade to other organisms, they help prevent erosion and are involved in nutrient cycling. Pine trees also represent an important resource for people, providing oxygen and timber.

Pine trees are woody perennial plants and around 200 species are known that extend across the northern hemisphere. Unlike other plants, trees contain a vascular cambium layer around the heartwood and sapwood, both of which are dead tissues. With all tissues inside the cambium being dead tissue, the mass of trees can be very misleading; the living layer may be less than 1 cm thick. The xylem and phloem tissues are responsible for water and nutrient transport and provide the link between the roots of the tree and the pine needles. Tree rings provide a marker of the growing seasons. When water is less available then the xylem and phloem tissues are thinner leading to darker rings in drier seasons. An important component of the project is therefore to define the living component of a pine tree.

The root system of a pine tree is both very complex and very large, comprised of tap roots and much smaller roots that respond to multiple stimuli in order to grow in the most favourable direction and conditions. The roots represent the tissues that are responsible for the intake of water and nutrients, but it is very small components of the roots (root hairs) that are responsible for intake of nutrients. There is therefore a contrast between very large and very small components. In the spring, roots spread out and sense where to grow by responding to gravitational, nutrient, and pressure stimuli. The roots then divide more and spread out in the more favourable directions. Therefore, whilst animals tend to have quite a defined geometry, plants are continually changing in terms of geometry and living mass. They are however confined in space, unlike many animals. Whilst pine trees have defined organs, these are not within a well-defined space and tend to be around and within other organs. The lifespan of pine trees is significant compared with other biota.

The ICRP model of a pine tree is comprised of an ellipsoid on top of a cylinder with an assumed uniform distribution of radionuclides across the mass of the organism. However, surface area will vary considerably for different tissues and these are not well represented using an ellipsoid. The roots are also excluded from the ICRP model and yet are a large and critical system for the trees, containing between 10 and 50% of the total tree mass. The roots are also largely concentrated in the top layers of soil so can be rapidly exposed to radionuclides deposited on soils from aerial deposition or from falling leaf litter.

The representative pine tree species selected for study as part of the project include:

- Japanese red and white pine;
- Scots pine; and
- Loblolly pine.

A literature review on growth rates between the different trees has been undertaken and a greenhouse trial established to compare growth rates and develop growth-mass relationships. Further work will involve taking CT scans of the trees to inform voxel phantom model development. Ideally a five year old tree will be selected for scanning, but this may be constrained by what will realistically fit within a scanner. There is also the possibility that there may not be enough contrast to work with the root system using young trees and magnetic resonance or neutron imaging may therefore be needed.

Further work planned involves visiting sampling sites at the SRS where soil samples are readily available along a pollutant gradient. The aim is to take tree cores that will be analysed for elemental content in relation to tree rings and compare against soil concentrations.

In terms of timescale, the aim is to complete a model for a living tree that includes the root system by the end of the winter (2015) and to include with data from the Savannah River tree core sampling work. In the future, the project may extend to focus on some of the smaller structures such as root hair cells and pine needle stomata, which are some of the more critical cells for tree functioning, and consider how to incorporate these tissues into the larger voxel model. One approach noted as potentially useful in this regard would be the use of positron emission tomography and K-40 labelling. Alternatively, since roots are correlated with soil hydrology, soil-penetrating radar or electromagnetic surveys could be undertaken to derive root density in association with soil hydrology.

Discussion

It was noted that information on trachea and bronchial systems and the mathematical techniques that can be used to represent these may be available in the Respiratory Morphology journal. This information may be relevant in terms of addressing the complex branching root system of pine trees. The use of data on imaging mechanisms used to derive leaf area index data, such as 3D laser scanning, may also be relevant for the canopy of pine trees. Useful literature would likely be available also on this topic (canopy/tree morphology). In general, scaling/fractal representation may be a useful alternative to detailed scanning or modelling of entire trees as, for example, there is a pattern how the branches attach to larger ones and how the needle structure organises itself.

Whilst the inner of a tree trunk is acknowledged as being dead tissue, under prolonged exposure conditions, this dead tree material will have had radionuclides deposited and these radionuclides will contribute to the exposure of the current living tissue. How to address this issue may warrant further consideration.

In combining dosimetry with voxel phantoms the suggestion was made to look at external dosimetry by considering experimental studies on Co-60 that were performed in the 1960's that looked at the effects of radiation exposure on forests as a function of distance from an elevated Co-60 source. The spatial distribution of observed effects and how the voxel model compares against this as a function of distance could be investigated.

2.11 UPDATE OF THE C-14 MODEL APPROACH FOR EL CABRIL VLLW AND ILW DISPOSAL…OR NOT?

Danyl Perez-Sanchez presented.

In Spain, disposal facilities for very low level waste (VLLW) and low and intermediate level waste (L/ILW) are operational at El Cabril (Figure 18). The models used for the biosphere safety assessment of the facilities are currently being reviewed and updated to take account of new knowledge on the safety assessment approach, to revise assessment scenarios to take account of river pathways and human intrusion, and to take account of special radionuclides, including C-14. The models will also be updated in terms of human land use and dietary habit assumptions.

The VLLW disposal concept was established in 2009 and differs from that for LLW in order to take account of the different waste forms. Contamination of infiltrating water will be measured for the disposal site.

Figure 18. The El Cabril facility for L/ILW and VLLW disposal.

The general scheme of the assessment is illustrated in Figure 19. In terms of the biosphere, the geosphere-biosphere interface is a stream system which then flows into a river that is used by people and agricultural animals, plus a range of other scenarios including accidents that include aircraft crash and the rising of the water table. Human intrusion is considered, but only after the point at which monitoring of the site ceases, some 300 years post-closure. The scenarios for human intrusion include construction and residential scenarios plus a residential agricultural scenario.

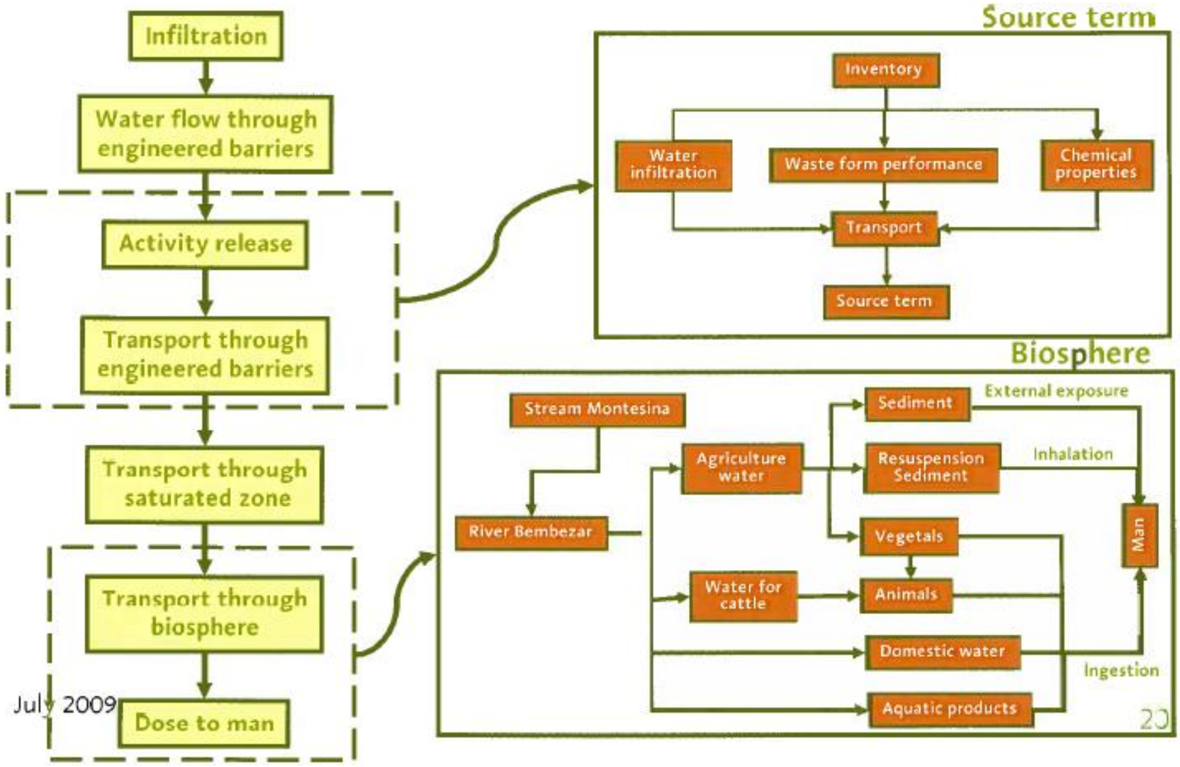


Figure 19. General scheme of the biosphere safety assessment.

The conceptual model for the groundwater pathway considers three aspects of source term analysis; water flow, activity release and transport through engineered barriers. Radionuclide migration to the biosphere and transport of radionuclides in the biosphere are also considered. A homogenous distribution of activity throughout the repository is assumed and the geosphere is conservatively assumed to be fully saturated. Two separate time intervals are considered – 0 to 300 years post-closure over which monitoring of the site will continue and >300 years.

In terms of C-14, the assumed inventory associated with L/ILW disposal is 20 TBq whereas for VLLW disposal the inventory is 0.2 TBq.

The biosphere stream-river scenario is focussed on La Montesina stream that flows into the Bembezar River. The stream can be entirely dry at some times of the year. In the surrounding area within 10 km, there are only 35 inhabitants. The largest town in the region is Homachuelo, which is located 31 km from the site and has a population of 3,000. This town is however located upstream of the stream discharge location and the part of the river that could become contaminated.

A range of different processes and pathways is considered in the stream-river scenario, including dilution in freshwater, use of water for human and agricultural animal consumption, transfer to fish and crustaceans, ingestion of contaminated food products by people and inhalation and external irradiation. The current model for dilution in freshwater is relatively simple, being based on average flux of river water and taking into account loss to river-bed sediment and re-suspension.

ENRESA is now giving more consideration to the biosphere in assessments than in the recent past. For C-14 in air, the RESRAD model has been applied, which takes account only of re-suspension as a mechanism for C-14 transport to air; gaseous release and photosynthesis are not considered. A general radionuclide model is not appropriate for C-14 and alternative models are being considered including specific activity approaches and more process orientated approaches that take into account transfer and interception factors. The two modelling approaches can also be used for evaluating C-14 concentrations in fish and crustaceans.

Assessed annual dose rates for the L/ILW facility are shown in Figure 20. Results indicate that C-14 is an important contributor to dose, but is by no means the most important radionuclide.

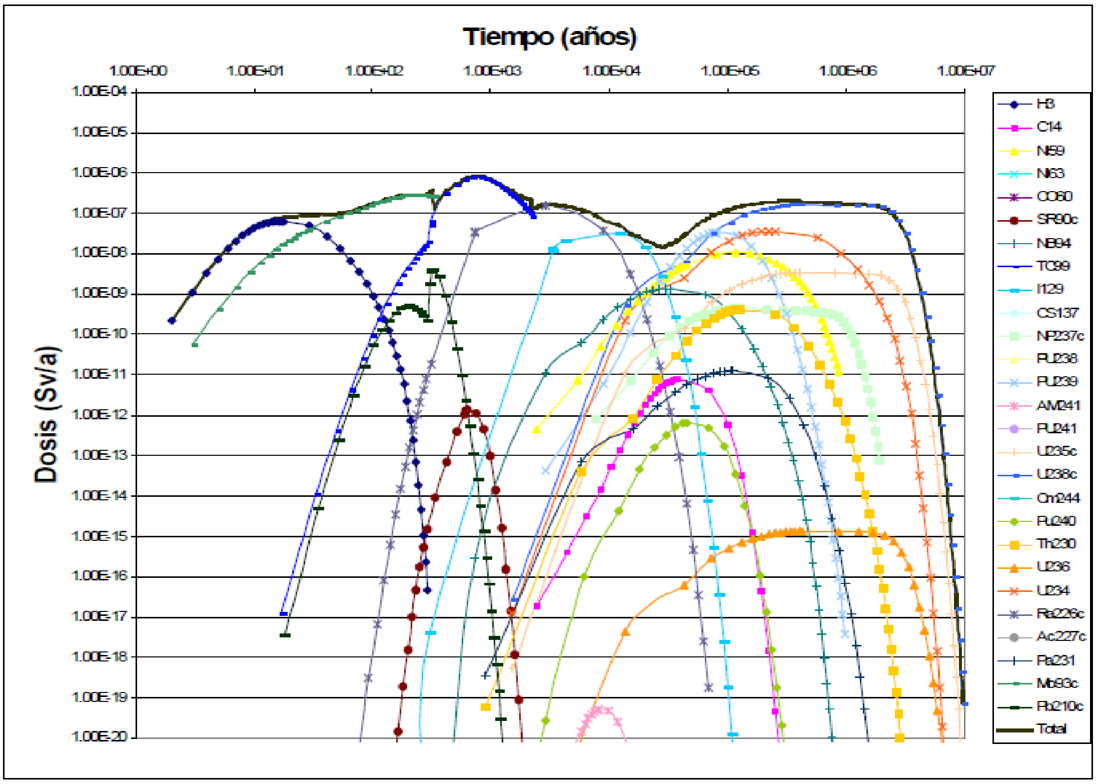


Figure 20. **Safety assessment results for El Cabril L/ILW disposal facility.**

The degree to which the C-14 model needs to be further developed is therefore being questioned in terms of whether additional processes need to be taken into account. There has to be a balance between model complexity and the availability of data for model parameterisation. Questions arise as to how to address transfers across the groundwater interface. For example, if the groundwater level reduces, will the radionuclides drop along with the groundwater or will they remain in pore spaces. If radionuclides do remain in pore spaces the question then arises as to whether they remain in place until groundwater rises once more or whether they volatilise. Redox kinetics can vary from hours to days and the oxidation state does not track instantly with the soil hydrology. The evaluation of radionuclide behaviour in soils under different hydrological regimes is therefore complex.

2.12 CARBON BALANCES IN WETLANDS

Ari Ikonen presented.

There are a lot of publications that describe mires and wetlands and a selection of available literature has been reviewed with the objective of working toward a conceptual model for such systems, particularly with regard to carbon cycling and C-14 dose assessment. A key characteristic of such systems is that they accumulate organic matter, but accumulation and loss rates vary considerably. Events such as wildfires and human perturbations such as drainage to allow for agriculture and forestry can have a large effect on mires. Furthermore, if drained, mires can become source of greenhouse gasses and of carbon (and other elements) to downstream waterbodies.

Plant communities in mires are governed by the microtopography. In dry areas hummocks can form above the water table that can host dwarf ericaceous shrubs and occasional trees. Where trees are present they tend to dry soils which results in more compact peat. The presence of dwarf shrubs with woody stems provides support to moss layers and also helps to maintain non-water logged conditions that allow other non-water loving plants to inhabit the area. At the water table, lawns can form that are characterised by linear-leaved plants such as cotton grasses and sedges. The roots of these plants can penetrate to some depth and are associated with intercellular gas spaces that help to protect against waterlogging. This plant community can produce and deposit organic matter into several peat layers also at a considerable depth. Sphagnum moss tends to form the matrix that supports growth of other species in both hummocks and lawns.

The wettest areas of mires are pools and hollows that are characterised by sparse macrophyte vegetation such as bog-bean and tend to be more similar to freshwater systems than terrestrial systems. Mires can support diverse fauna at all trophic levels across hummocks, lawns and pools/hollows.

The top few centimetres of moss are densely packed, preventing lower layers from receiving light which leads to death and decay in these lower levels. The moss mass is highly permeable to gas and water so degradation is mainly aerobic and DOC and carbon dioxide are produced. However, as the moss continues to grow upward, the mass is pressed down and compressed so the lower areas are in a constant state of collapse which ultimately results in peat formation. The porosity decreases as air spaces are reduced and the hydraulic resistance is increased. This leads to more horizontal water fluxes. The formation of peat in mires is a constant process. Within the top few centimetres of the waterlogged zone, oxygen is consumed by microbes faster than it can be replaced from air and the peat becomes anoxic. This results in further decay although the rate is much slower and is driven by bacterial action. Here it is mainly DOC and methane that are produced. Fermentation through sulphate reduction may also be important.

Gas production occurs throughout the full depth of a mire. Movement may be by diffusion, ebullition as bubbles, or through transport through gas spaces in plant roots and stems in the surface layers whereas within the deeper layers it is largely diffusion that governs gas transport. Gases and DOC have been observed to be younger than the surrounding peat, suggesting a general downward transport of carrier pore water.

The escape of gas from mires can be variable. For example, horizontal transport and release from hummocks could occur. Alternatively there may be direct release from wetter areas of the mire. The reviewed literature suggests that up to 80% of the methane in mires would be oxidised to carbon dioxide in the upper oxic layers of the mire prior to release to atmosphere. The redox interface where the microbial community is most active may therefore be a key area governing carbon behaviour.

Eventually equilibrium will occur where peat accumulation equals production and decay due to the limited growth of the plants. However, the timescale for equilibrium to occur can be long; peat bogs in the boreal regions have continued to grow for the last 10,000 years.

There are a few papers that have focussed on open water pools, but carbon cycling in pools can be expected to be comparable to other water bodies, although biota communities are very different. The pools contribute less to peat generation, but may give rise to higher methane production or a faster transport path to the surface/atmosphere. On a small spatial scale there is wide diversity in the surface of mires with dry and wet areas varying considerably, which leads to a large variability in productivity.

Mire pools can be considered as multiple small pools or as larger inter-connected areas. Mire pools are common, but have not been well studied to date. The pools tend to be quite shallow so there is a large surface area to depth ratio, which can result in relatively large losses of methane etc. The pools contain lots of organic carbon sourced from the surrounding peat. The pH can vary considerably depending upon whether there is groundwater input to the pools or not. The pools can be 'hot spots' for fauna due to algal primary production, the presence of detritivores and the microbial cycle. The main primary productivity is associated with phytoplankton. A wider range of invertebrates is associated with the pools as compared with peat. Molluscs tend to be excluded however since there is insufficient calcium present for shell growth. The importance of pool areas to birds and mammals is not so well known, but the areas are known to be important for amphibians. The pools therefore have quite distinctive food webs. In winter, the top layers of pools may be frozen but lower layers remain active with plankton production potentially occurring on the underside of ice where light penetrates. Rates of productivity will however be greatly reduced.

If C-14 enters a mire, it would be difficult to evaluate where the input would occur due to the mosaic nature of these areas. Larger mires typically form on former lake basins in boreal regions. These lake basins are often associated with clay layers which may result in groundwater moving laterally below the clay. Input may therefore be from the borders of the mire. Preferential flow routes may also occur associated with fractures in the bulk of the peat layers.

Mires are characteristically sinks for both carbon and water, although outlets can exist. Losses of carbon may also occur as a result of flooding events. The age of a mire governs both the arrival routes and transport and release for C-14. As peat grows, central areas may become ombrotrophic. Open pools may remain to link the mire to groundwater or may be severed as the peat bog rises. The source of C-14 to a mire may not be limited to groundwater input, surface deposition may also occur following the uptake and release of C-14 by plants as well as transport from the upstream areas. Where C-14 is present in mires, consideration would need to be given as to the source (e.g. groundwater input) as opposed to the C-14 baseline from when the peat formed.

For tree covered areas, Lagrangian models are required to represent averaged ventilation fluxes. More advanced modelling is however required to represent what is happening below the canopy of the trees. The Atmosphere-Plant Exchange Simulator (APES) is a good example of such a model as this is a multi-species, multi-layer process model.

Available data on mires tend to be from different mires, hence it is difficult to quantify a universal picture of carbon cycling. Nonetheless, most of the biomass of a mire is associated with the peat and moss matrix. Mire pools host only a small amount of the carbon that cycles through a mire system, but they are potentially important due to the biodiversity of such pools and the specific biota communities they host; such pools can host specialist and rare/endangered species.

2.13 TRANSPORT OF C-14 IN AGRICULTURAL ECOSYSTEMS: REMAINING ISSUES AND UNCERTAINTIES

Mike Thorne presented.

As illustrated previously (see Figure 4), there is a range of processes associated with carbon (and hence C-14) behaviour in agricultural systems that operate over varied timeframes and some calculations have been performed to represent these processes, in line with observations from the RWM field experiments.

Methane in the atmosphere diffuses into soil where it is gradually reduced in concentration with soil depth. If the flux into soil and methane concentrations in air are considered, the ratio of concentrations is related to the length scale over which consumption of methane occurs in the soil. This has been represented using a first order reaction rate constant. Where this is inverted, a length scale over which methane is metabolised is produced. This equation has been applied to antecedent head space measurements obtained from the RWM field experiments. The resultant length scales were all around 0.1m. Similar results were obtained from data arising from the laboratory studies. Methane oxidation rate constants were also calculated and good similarity was again observed.

If the flux from the soil to the head chamber is ignored and instead the soil profile is focussed upon, longer length scales are calculated and the oxidation rate constant for methane is around an order of magnitude lower. The same equation was applied to investigate that percentage of methane injected at depth that is emitted from the soil surface. Of the methane injected at a depth of 50 cm, around 23% is emitted as a result of methane diffusion. The majority of the injected methane is converted to carbon dioxide.

A detailed model has been developed on the basis of the results of the RWM studies. The model is based on the profile of gas concentrations through the soil and efflux from the soil. The concept is that methane diffuses away from the point of injection and microbes then convert this into carbon dioxide. The carbon dioxide then diffuses upward through a partially saturated soil. The key processes are therefore the diffusion of gasses through partially saturated soil, microbial activity and soil respiration, which is the degradation of soil organic matter to produce carbon dioxide which itself affects the background concentration profile of CO_2 in the soil.

The developed model (T2Plants) is a sophisticated computational model that accounts for all of the key processes and also accounts for isotopic effects. The model is based on TOUGH2v2 which is a computer program for simulating non-isothermal flows of multi-component, multiphase fluids in porous media. The option to couple the model to surface atmosphere boundary conditions is provided for since the boundary conditions can drive the system. The influence of plants on the system was also considered. The model, illustrating $^{12}CH_4$ through a soil column, is presented in Figure 21.

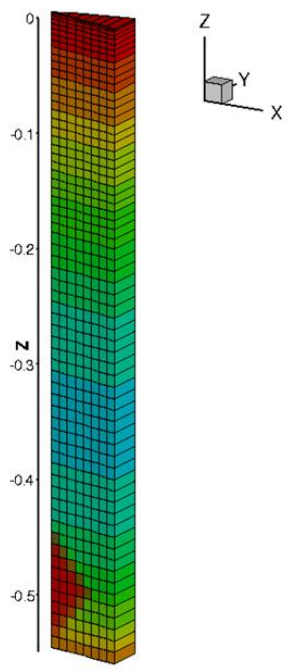


Figure 21. Model representation of a soil column showing $^{12}CH_4$ through the column.

The model has been calibrated against the fourth laboratory experiment results, focussing on the first hour of the experiment, and has then been applied to predict further time steps. Small kinks in modelled curves are observed (Figure 22) which relate to the extraction of soil gas from different depths of the soil column. Results indicate that the sampling regime is not a major perturbation on the modelled system.

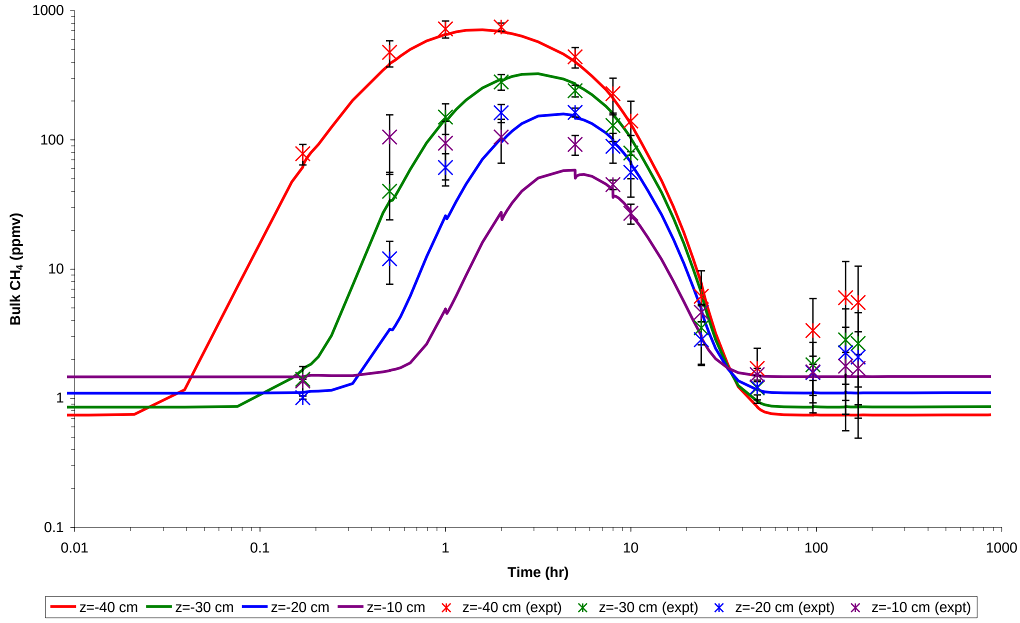


Figure 22. Model prediction of average bulk methane concentrations at different depths of a soil column over time.

Blind predictions of CO_2 concentrations in field experiment 2 and laboratory experiment 4 were also made, assuming that production of carbon dioxide is uniform throughout the soil column. The model simulation of the carbon dioxide profile, which balances production rate and diffusion, is very good for the laboratory column experiment. However, for the field experiment, the model overestimates carbon dioxide at depth, but is more accurate in the higher soil zones. At around 40cm depth there is a change in the soil texture and organic matter is present mainly in this zone. The model however assumes a uniform organic matter profile and this is likely to explain the over prediction. A delay in carbon dioxide production is also observed from the experimental results. This is due to the conversion of methane to carbon dioxide via formaldehyde, a multi-step process.

The antecedent profile of methane in field experiment 2 has also been computed, with the model results suggesting consumption at a faster rate than observed in the experiment, suggesting over estimation of the k value. The model has to handle bulk methane concentrations at 40cm that reduce with diffusion up the soil column and was found to be good at picking up peaks in methane concentrations at intermediate depths and release from the soil surface. The model is quite constrained with only a couple of parameters that can be varied for calibration.

An anomaly has been observed with regard to the isotopic profile, which suggests that C-13 methane is being oxidised at a greater rate than C-12 methane. In a simple system, isotopic effects more than a few percent would not be expected. However, this is not a simple system; porosity will vary throughout the soil. At the injection site there is a greater porosity, which may lead to diffusive discrimination that is emphasised by the different scales of diffusion throughout the soil. The concentration of bulk methane drops considerably through the soil and it is the residual few percent remaining within the soil column in which the isotopic anomaly is observed, with 4% of the heavier and 2% of the lighter methane remaining. The diffusion of methane through the soil occurs at the expected rate. A time lag was observed between the consumption of C-13 methane and the formation of C-13 carbon dioxide.

There are several different models available to represent the transport of C-14 through soil. However, how processes are handled varies in the models. The way in which the plant canopy is represented has also evolved from a very simple representation to a more complex one.

In the initial version of the assessment model that has been developed, a complete oxidation hypothesis was adopted that was found to be conservative by around 25% in the context in which it was applied. In the second version of the model, a length scale was applied with a modifying factor to account for tortuosity and variation in soil moisture with depth; as soil is filled with water there is a reduction in the volume of the air filled spaces that allow diffusion leading to a non-linear diffusion profile. The tortuosity model has been calibrated over a wide range of soil types.

In terms of uptake into plants, there are two boundary conditions of particular interest – the above canopy boundary and the below boundary condition (i.e. the flux from soil). The concentration in the above-canopy atmosphere is calculated using dispersion theory and depends upon the size of the release area. If there is a small release area and a unit flux then there will be a small concentration, but if there is a larger area with same flux the concentration will be greater. The flux from the soil is independent of the area of release. The upper boundary condition is therefore proportional to fetch with the lower boundary condition being independent of fetch. Within the canopy the influence of area will vary somewhere between the two. If the bottom boundary condition dominates then the concentration in the sub-canopy will be independent of area, whereas if the above boundary dominates then the sub-canopy concentration will be related to fetch. Which boundary is dominant needs to be considered on a case by case basis.

Once the sub-canopy concentration has been derived, the concentration in plants can be derived by equating the specific activity in the sub-canopy atmosphere to that in plants. Whilst there will be subtle differences observed for different phases of the growing season, there will not be enough variation to argue for such a detailed level of consideration in assessment models and the focus can therefore be on the total plant concentration.

The plant concentration resulting from sub-canopy uptake can be augmented by the uptake of dissolved carbon dioxide in soil water by plant roots. To calculate uptake, the water requirement of plants over the growing season, expressed as a transpiration requirement and often around 500 l of water per kg of plant, is required. The water is assumed to be transpired by leaves, but the C-14 in carbon dioxide is captured for photosynthesis. If it is assumed that half of this captured C-14 is used in respiration rather than biomass production, a value of around 0.6% is derived for the amount of C-14 derived from root uptake for a typical crop. This value could however be as high as 2% for some crops.

Once plant concentrations have been calculated, a specific activity approach is used to derive the C-14 concentration in animals and humans.

The main uncertainties in the assessment model relate to the concentration of C-14 carbon dioxide in the plant canopy atmosphere. These uncertainties arise from the necessary simplification in the description of the canopy and on the dispersion model used to describe processes above and within the canopy. The model assumes small plants with stable atmospheric conditions and a low wind speed, which probably constitutes a cautious set of assumptions.

There are fewer uncertainties associated with the soil model. We have a good handle on diffusability and tortuosity under different soil moisture conditions in agricultural soils and the length scale for conversion of methane to carbon dioxide has been calculated as a few tens of centimetres. As such, provided the water table is at depth (more than a few times the length scale), conversion will be essentially complete, regardless of the particular values of the diffusion coefficient and the oxidation rate constant.

A further uncertainty relates to the dose resulting from the ingestion of contaminated plants, which relates to the fraction of carbon in a person's diet that could be obtained from locally sourced foods. The model cautiously assumes a fraction of 0.3.

Beyond the assessment model itself there remain further uncertainties, not least of which is the release of other gases from a disposal facility, such as carbon monoxide. There is some evidence that the oxidation rates for carbon monoxide in soils can vary considerably and the reasons for this variation are not well understood. Where CO is released to air, it can be utilised by plants and incorporated into sucrose and proteins rather. In darkness, carbon monoxide is absorbed at close to the same rate as in light, but is almost completely converted to carbon dioxide and released as the conversion process for sucrose and proteins is turned off. During daylight, the rate of fixation appears to be proportional to the concentration of carbon monoxide, but is unrelated to the rate of photosynthesis. The fixing capacity for vegetation has been calculated as around an order of magnitude more rapid than the rate found in soils at higher concentrations of carbon monoxide. Plants are therefore very efficient at sequestering carbon monoxide from the air. Carbon monoxide released to air therefore goes straight into plants whereas methane released to air must first enter the soil and be oxidised to carbon dioxide before it can be taken up by plants.

In terms of effective dose factors (Table 6), the lowest dose factor is for methane due to a prolonged and indirect process. The dose factor for C-14 in CO_2 will be higher since the C-14 can then go directly into a plant and be used in photosynthesis. The dose factor for C-14 in CO will however be higher still since this goes straight into metabolites rather than relying on the photosynthetic system. Previously carbon dioxide has been considered as the worst case atmospheric discharge from stacks, but consideration needs to be given as to whether we can justify an assumption of 100% release as CO_2 rather than CO. Where carbon monoxide is assumed to be released, its behaviour in the atmosphere then needs to be considered. It may be that carbon monoxide can be oxidised to carbon dioxide, but the fact that carbon monoxide poisoning occurs suggests that this conversion is not instantaneous.

Table 6. Effective dose factors (Sv y^{-1} per Bq y^{-1}) for different chemical forms of C-14 released at a height of 15 m.

Age Group	Carbon dioxide	Carbon monoxide	Methane
Infant	$9.0\ 10^{-18}$	$1.2\ 10^{-16}$	$1.5\ 10^{-20}$
Child	$5.5\ 10^{-18}$	$7.1\ 10^{-17}$	$8.7\ 10^{-21}$
Adult	$5.5\ 10^{-18}$	$7.2\ 10^{-17}$	$9.0\ 10^{-21}$

One factor that has not yet been considered in the RWM assessments for C-14 is what happens when the release ceases. The continued input of C-14 to plants will be governed by organic matter turnover in soils and, as such, residual carbon pools need to be evaluated. There are some models that consider organic matter retardation in mineral soils, but these may not cover all situations. Furthermore, currently it is presumed that C-14 uptake by roots is a passive process, even though subsequent utilisation is active. It has yet to be established if uptake really is a passive process. If organic carbon compounds are present in soils, there is a possibility for active transport by roots. The question of root uptake therefore remains unanswered. Nonetheless, a passive uptake assumptions is adequate in explaining observations from experiments. It is not yet known whether C-14 entering leaves from the transpiration stream is available for photosynthesis, but the assumption that it is so available is cautious.

2.14 STATUS OF THE REVISION OF IAEA SRS-19: APPLICATION OF SIMPLIFYING ASSUMPTIONS AND A GRADED APPROACH IN PROSPECTIVE SCREENING OF RADIOLOGICAL DOSES FOR PLANNED EXPOSURE SITUATIONS

Graham Smith presented on behalf of Tamara Yankovich.

The IAEA is currently running a program to update the document 'Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment, Safety Report Series 19 (2001)'. The update will allow for screening assessments to be undertaken within a graded approach for both people and the environment. As such, the update will include three volumes. Volumes 1 and 2 will focus on human protection, with volume 1 covering screening assessments of public exposure and volume 2 providing details on the generic models that underpin those assessments, including details of assumptions employed and parameter values. Volume 3 focusses on plant and animal scenarios for planned exposure situations. The objective with volume 3 is to facilitate the application of screening models to biota, ensuring that there is some level of confidence in the assessed implied risk. The screening approach will be based around the ICRP Reference Animal and Plant approach, taking into account the latest information on environmental transfers.

The update is not intended to be applied to emergency planning/response scenarios or to long-term assessments for radioactive waste disposal. The scope will however include C-14 in sewage sludge and disposal in landfill for which information and knowledge exchange may be beneficial.

The SRS-19 update will fall into the category of IAEA guidance, providing a structured approach to prospective dose assessment from simple screening assessment through to more detailed assessments. A particular emphasis is on ease of use. The update will consider 122 radionuclides in volumes 1 and 2 (assessments for people) whereas fewer radionuclides will be covered for biota dose assessment due to the lack of transfer data currently available. A key future consideration will be to identify which data gaps are important and require filling.

The update will provide tables of environmental dilution factors for discharges to air and water, in Bq/m^3 per Bq/a discharged. (In some ways these correspond to the GBI for solid waste disposal since this is where dilution could occur, for example, in meteoric near-surface water bodies). Screening coefficients will also be provided in terms of dose rate per unit discharge rate and dose rate per unit activity concentration.

The assessment models are required to be conservative, but not unreasonably so. Furthermore, a sound scientific basis for assessments is required, which includes developing a good understanding of the underlying transfer and exposure processes. The key questions being considered during the update of SRS-19 are:

- Can a graded or risk-based approach be applied, such that the degree of priority and level of effort are commensurate with risk?
- What level of conservatism would be required to ensure protection, such that effort is commensurate with risk?

- How conservative should model assumptions be? and
- What should be measured?

Five categories of assessment are being considered (Table 7). Category one is a simple screening assessment whereas category five relates to very detailed site-specific assessments relating to nuclear power plants and other large nuclear installations. Whilst information within the revised SRS-19 will be helpful for such assessments, site-specific models will be required.

Table 7. Categories of assessment in the revised SRS-19.

Category 1	Category 2	Category 3	Category 4	Category 5
One-step assessment No dilution of the discharges No site-specific information necessary Discharges to air or to rivers	Two-step assessment Includes dispersion in the environment Limited site-specific information is required Discharges to air or to rivers	Includes dispersion in the environment Customisable to include site-specific information and individual exposure pathways Discharges to air or to any surface waters	More detailed estimation of dispersion in the environment Customisable to include site-specific information and individual exposure pathways Discharges to air or to any surface waters	Detailed assessment Includes dispersion in the environment Customisable to include site-specific information and individual exposure pathways Discharges to air or to any surface waters Uncertainty can be specifically quantified

A range of key issues is still being worked on, including justifying the application of the 75th percentile for input parameters. It was noted that, in the UK, it is commonly either the 90th or 95th percentile that is applied with the 75th percentile not being recognised as a standard approach for parameter value selection. Work is ongoing within the IAEA to check that assumptions provide adequate conservatism.

Finally, it was noted that, in addition to the SRS-19 update, the IAEA is developing a Technical Report Series document on ‘Uranium transfer in the environment’ for which uranium data are being sought to support the document development. The TRS will consider, among others, NORM and mining issues relating to uranium and will consider both modes of action (radiotoxicity and chemotoxicity) and impacts on people and the environment. Further documents are due to be developed on polonium, radium and lead.

3. DISCUSSION AND RECOMMENDATIONS

In response to the presentations made by participants, as reported in Section 2 of this report, participants were invited to discuss those aspects of C-14 dose assessment considered to be key to addressing remaining uncertainties and whether there would be scope for further collaborative work to address these remaining uncertainties and/or improve confidence in assessment models. The remaining uncertainties are discussed in Section 3.1. Section 3.2 then provides a summary of discussion around future collaborative opportunities. With interest being expressed by participants of the workshop on a number of possible collaborative areas, ideas for future activities were agreed and responsibilities identified. These are detailed in Section 3.3.

3.1 REMAINING UNCERTAINTIES IN C-14 DOSE ASSESSMENTS

In terms of source terms, carbon monoxide has been identified as a potentially important issue that may require some further consideration; it was noted to be an issue in the near-field and, if this can escape it could then become an issue for the biosphere. Coherence of assumptions for behaviour in the near-field, transfer to the biosphere and behaviour in the biosphere would require review and comment by others.

The issue of carbon monoxide was raised in an operational dose assessment undertaken by RWM that included a literature review of the subject. It was suggested that those interested in the C-14 issue could review the discussion around carbon monoxide and consider whether it is of relevance to their own assessments. How carbon monoxide behaves in the soil-plant system may be a question that could be addressed by the IUR FORUM.

The endpoints of interest in C-14 assessments may differ between assessment communities (e.g. carbon fluxes as opposed to C-14 doses), there is scope for sharing experience and understanding of the different terrestrial and aquatic systems. At low doses and dose rates, the micro distribution of C-14 (a low energy beta emitter) in organisms may be important, particularly for organic forms. It is unlikely that the exposure resulting from the micro-distribution will be important in terms of dose assessments, but the issue should be thought through for the range of organic compounds that may be associated with C-14.

Whilst the timeframes of interest for assessment communities may differ, there is a need for short-term processes to be understood in order to justify simplifications for long-term assessment models and to determine which carbon pools are in equilibrium so as to ensure that temporal and spatial averaging scales are appropriate. In this regard, there is also an issue around how to connect short term processes within a long-term model and similarly how to apply data from short term models in an appropriate manner within a long-term assessment. Assumptions and/or abstraction will be required in moving from short time step data to the parameterisation of a long term model. This may require parallel model development with one model focussing on detailed modelling of processes and another being used to represent the results of detailed modelling for long-term assessment. Such a process can help to ensure that simple models are conservative, but not overly so. Guidance on relevant averaging of data values to represent processes in the long-term would nonetheless be helpful. For example, in terms of photosynthesis, the arithmetic mean of photosynthetic rate over a 24 period may not be appropriate since rates will vary considerably with light availability. Incorrect averaging could have a real impact on the assessed doses.

A recent paper following an IAEA model inter-comparison programme illustrated the need to improve models for assessing doses to non-human biota in wetlands; orders of magnitude differences in assessed exposures were evident between models applied to a wetland scenario[k]. A NERC funded project was noted to have begun to look at the issues in more detail. Further work programmes have been established within COMET and TREE to look further at the impacts of radiation on non-human biota and to improve assessment capabilities.

Related specifically to C-14 in biota, it was noted that accumulation of carbon tends to be more in muscle than in fatty tissues although, in some shelled organisms such as mussels, C-14 largely goes to the shell rather than the soft tissues.

The ecosystems that are particular priorities in assessments and for which research activities are undertaken can be driven by particular geological and political contexts. For example, in the UK, the regulatory regime makes it likely that a repository for ILW would be situated in a lowland area which, in the UK, largely implies agricultural systems. Bulk hydrogen and methane release are a concern for a L/ILW repository and this was largely a driver for the field experiments that were conducted on behalf of RWM, in order to gain a greater understanding of processes that would affect human doses. For example, in Finland and Sweden, wetlands are a particular focus due to their likelihood as discharge areas. A particular interest, in the context of landscape evolution, is what happens to C-14 that discharges to a mire when that mire becomes something else (e.g. through drainage for use as agricultural land). Mires themselves are nonetheless of interest due to the potential for non-human biota exposures.

Grasslands have been studies by IRSN and ryegrass and spring wheat were studied in laboratory and field experiments undertaken on behalf of RWM to evaluate aspects of C-14 behaviour, but behaviour in relation to other crops has not been studied to date. Different crops would have different root systems that will affect C-14 diffusion through soils. A balance needs to be struck between studying a sufficient number of crops to gain a scientific understanding of behaviour in relation to different root structures and making reasoned interpretation and interpolation to allow a judgement to be made on the applicability of a model to different root systems.

A special model approach is required for C-14, but also for other nutrient elements. Within BIOPROTA, agricultural canopy processes have been considered, but the same level of detail has not been given to understanding processes in wetlands, forests or in freshwater. Wetlands in particular could provide microsites with strong changes in redox conditions that could have a large effect on C-14 behaviour and both methane production and consumption could be observed in the same soil. There also remain sub-surface transport issues that have not yet been resolved and where volatilisation effects can occur.

Finally, it was noted that, in the UK, radionuclides are currently being formally assigned to the class of ‘hazardous substances’ in terms of the Groundwater Daughter Directive. The Directive requires that all hazardous substances should be prevented from entering groundwater, but the definition of ‘prevent’ is being actively discussed. What is considered as groundwater is also being discussed to determine whether this relates to all water below surface or whether the focus should be on groundwater aquifers. The approaches being taken in other countries to address the issues raised by the Directive would be of interest in informing discussions.

[k] Stark K , Andersson P, Beresford N A, Yankovich T, Wood M W, Johansen M P, Vives i Batlle J, Twining J,. Keum D-K,, Bollhoefer A, Doering C, Ryan B, Grzechnik M and Vandenhove H (2015). Predicting exposure of wildlife in radionuclide contaminated wetland ecosystems. Environmental Pollution 196 (2015) 201-213.

3.2 FUTURE COLLABORATIVE OPPORTUNITIES

There are a number of other communities interested in carbon that may provide opportunities for sharing knowledge, including carbon dioxide geo-storage and climate change research communities. Those involved in town gas storage (methane and hydrogen) are required to consider what happens to these gasses under geo-storage conditions and what would be the consequences of any leaks. As such, relevant data for long-term assessments may be available. Similarly, the climate change community may have relevant information with regard to organic carbon in the near surface and on the cycling of carbon in peat and mires (as well as mineral-soil forests) through research into whether or not carbon would be released from these systems under changing climate conditions. More detailed understanding of climate impacts on carbon cycling was recognised as an important requirement for long-term C-14 dose assessments. In support of this, a long-term experiment on persistent organic matter in soils would be of interest to investigate the important exchanges governing organic carbon behaviour in soils and uptake into plants and whether long-term storage in soils with subsequent release should be explicitly considered in assessment models. Such an experiment would be complimentary to the long-term studies that have been undertaken at Rothamsted, UK over more than 100 years.

The sharing of existing data sets to allow models to be compared, both against data and against other models is a very useful exercise in improving confidence in assessment models and the IRSN field data set from an agricultural field close to the La Hague reprocessing plant has provided an interesting basis for a model intercomparison study for IRSN and SSM. There was interest in extending the study to other modellers and IRSN are happy to make available the data set for this purpose. The RWM field data set also presents an opportunity for a scenario to be developed to which models can be applied and compared. In particular, this study could help to investigate whether root uptake is a relevant process to include in assessment models. Workshop participants were invited to review the output of the RWM studies to determine whether the interpretation of results seems appropriate and/or whether alternative interpretations can be made. Review of the model is also invited. The full derivation of all equations has been reported and participants were invited to scrutinise these and see whether there are assumptions that could be challenged. Application of other models to the experimental data set would be beneficial to this end. The data have already been formally requested from RWM by SSM.

Root uptake estimates have previously been between 1% and 5%, but the experiments from which these values were derived were at the limit of detection. Improvements would therefore be useful. It was noted that work has been undertaken by Keiko Tagami from NIRS in Japan on C-13/C-12 ratios in soils and plants in order to investigate root uptake based on this isotopic ratio. This approach could be applied to other crops to gain a greater understanding on the proportion of carbon derived from soil by root uptake and that from air by photosynthesis. It would also be interesting to enrich organic matter with C-13 to investigate root uptake in relation to organic matter.

In addition to the studies undertaken by IRSN and for RWM, C-14 data relevant to model testing for waste disposal may exist in relation to the AECL Duke Swamp site. A number of publications are available, in addition to the research reported herein. There may therefore be merit in evaluating available data, plus any further data that could be made available by AECL, to evaluate whether a further scenario can be developed, potentially incorporating a wetland system and investigation of processes at the interface between the geosphere and the biosphere. This could also provide a good opportunity to test the BIOPROTA GBI methodology.

Duke Swamp is a complex site however as it is a largely natural and a whole environment rather than being a constrained system as in the RWM and IRSN experimental systems. There are nonetheless measured C-14 data from upstream of the swamp and within the swamp so it would be possible to start asking process-based questions on the spatial distribution along and from the geosphere-biosphere boundary. A process model could then be considered that maps the distribution along that boundary and within the swamp. Supplementary data on tritium in the system could be used to help understand features of the system that could affect C-14 behaviour. Alternatively, the focus of an exercise could be on the swamp itself, without the linkage to the upstream, such that a detailed process model may not be required. A starting point would be to collate available data and use this to develop a conceptual model.

In addition to Duke Swamp, there is a large volume of literature on mires and wetlands relating to their characterisation and C-14 profiling. Collation of this data could provide useful information and data that could be used to improve confidence in assessment models of these systems. Further data that may be of interest, and that could be made available, has been identified from the Food Standards Agency (FSA) in the UK. The FSA has data relating to historical discharge monitoring for C-14 and discussions are underway to determine how this could be used in future work.

3.3 SUGGESTIONS FOR FUTURE ACTIVITIES AND IDENTIFIED RESPONSIBILITIES

There was much interest in extending the IRSN/SSM modelling exercise and Laura Limer (Quintessa) was noted to be in a good position to develop a proposal for this, based on the IRSN data set and in consultation with IRSN. A request has also been made by Quintessa for the RWM data set and she would therefore be in a good position to also consider a modelling exercise around these data. Quintessa was therefore tasked with taking forward the development of a proposal for C-14 modelling exercises framed around these data sets.

Duke Swamp was considered to present a separate opportunity that would begin with the collation and interpretation of available data and, from this to develop a conceptual model. The swamp also provides an opportunity for the BIOPROTA GBI methodology to be applied to a real site. Consideration for a programme around data at Duke Swamp continues and Mike Thorne was suggested as an appropriate person to take this forward in relation to the potential GBI methodology application.

Ari Ikonen (EnviroCase) expressed interest in developing a proposal around the identification and review of literature relating to mires and wetlands. There are well characterised mires in Finland, Sweden and Canada, among others, that could provide useful data in support of model testing. The work programme would therefore initially focus on identifying appropriate data sets that could feasibly be used to validate wetland models.

APPENDIX A. LIST OF PARTICIPANTS

Participant	Affiliation
Achim Albrecht	Andra, France
Ari Ikonen	EnviroCase, Finland
Beatriz Lourino-Cabana	EdF, France
Caitlin Condon	Oregon State University, USA
Danyl Perez-Sanchez	CIEMAT, Spain
Graham Smith	GMS Abingdon (BIOPROTA Technical Secretariat), UK
Jon Napier	Oregon State University, USA
Karen Smith	RadEcol Consulting (BIOPROTA Technical Secretariat), UK
Kirsi Riekki	Posiva Oy, Finland
Laura Limer	Quintessa, UK
Loïc Ducros	IRSN, France
Maria Nordén	SSM, Sweden
Mike Thorne	Mike Thorne and Associates, UK
Peter Saetre	SKB, Sweden
Séverine le Dizès-Maurel	IRSN, France