BIOPROTA

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

C-14 in the Biosphere

Report of an International Workshop

Laura Limer (Ed.)
Version 2.0, 8 October 2019

PREFACE

BIOPROTA is an international collaboration forum designed to support resolution of key issues in biosphere aspects of assessments of the long-term impact of contaminant releases associated with radioactive waste and contaminated land management. It is understood that there are radioecological and other data and information issues that are common to specific assessments required in many countries. Collaboration through projects focused on mutual research needs 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 and related assessments of radioactive waste management. Particular emphasis is placed on key data required for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge to the environment or release from solid waste disposal facilities. The 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 10-11 April 2019, hosted by IRSN in Aix-en-Provence, France. Technical input was provided by a range of organisations via presentations and discussions, as described in this report. The financial support for the workshop provided by Andra (France), Nagra (Switzerland), Posiva (Finland), SKB (Sweden), and SSM (Sweden); the support provided by IRSN (France) in hosting the workshop, is also acknowledged.

Version History

Version 1.0: First full draft compiled by Laura Limer based on input from participants at the workshop and reviewed by the Technical Support Team. Distributed to participants and BIOPROTA project sponsor organisations for comment on 25 June 2019.

Version 2.0: Updated following feedback from participants, finalised and distributed to BIOPROTA members and workshop participants on 8 October 2019.

CONTENTS

1. INTRODUCTION

C-14 can be a radionuclide of importance in assessing the safety of discharges to the environment from operating facilities (including nuclear power plants, existing waste disposal facilities, radiopharmaceutical production, hospitals and associated research facilities), historical facilities (including historical waste disposals) and potential future facilities (e.g. planned or proposed power plants and waste disposal facilities). As a radioisotope of a fundamental component of organic molecules, C-14 requires special consideration when assessing potential consequences of releases to the biosphere.

Over the past thirteen years, a sequence of BIOPROTA workshops, model-data and model-model comparison exercises have contributed to both improvements and helping to build confidence in the representation of C-14 in assessments covering both short and long-term releases to the biosphere.

1.1 AIMS AND OBJECTIVES OF THE 2019 WORKSHOP

The workshop aimed to support further improvement and harmonisation in the assessment of C-14 in the biosphere through sharing associated information, experience and knowledge, consistent with the BIOPROTA concept. In particular, the objectives of the workshop were to:

- ☐ present and discuss research and assessment modelling experience relating to C-14 in terrestrial and aquatic systems;
- ☐ review progress made in parallel with the BIOPROTA studies relating to C-14; and
- ☐ identify opportunities for building further confidence and/or further potential improvements in the representation of C-14 in biosphere assessments.

The outcome of the workshop is this summary of the presentations given, and an identification of topics where further collaboration would help to build confidence in C-14 modelling in the biosphere.

1.2 PARTICIPATION AND SUPPORT

The workshop was attended by 19 participants from 10 countries, representing a range of operators, regulators, researchers and technical support organisations. Participants and their organisations are listed in Appendix A.

The workshop was hosted by IRSN in Aix-en-Provence, France, 10-11 April 2019. Sponsorship was provided by Andra (France), Nagra (Switzerland), Posiva (Finland), SKB (Sweden), and SSM (Sweden) and the workshop was facilitated by a Technical Support Team, as listed in Appendix A.

1.3 REPORT STRUCTURE

This report provides a summary of the presentations and associated discussions in Section 2. Section 3 then provides an overview of the concluding discussions and of possible collaborative tasks that could be taken forward.

2. PARTICIPANT PRESENTATIONS AND DISCUSSIONS

Following participant introductions, an overview of previous BIOPROTA collaborative work and issues remaining was provided by the Technical Support Team. Presentations were then delivered by participants on developments in modelling approaches, assessments and potential data sets that may be considered for model testing. The presentations and associated discussions are summarised below.

2.1 REVIEW OF PREVIOUS COLLABORATIVE BIOPROTA WORK

Graham Smith (BIOPROTA C-14 project Technical Support Team) presented.

The presentation started with some background into C-14 biosphere assessments. Initial interest in a C-14 specific dose assessment arose in the late 1980s. For example, an assessment of the potential impacts of radioactive waste disposal in the near-surface disposal facility at the LLW Repository Ltd site in Drigg, UK, determined that the potential impacts were dominated by naturally occurring radionuclide decay chains and C-14 [Smith et al., 1988]. This insight has contributed to developments in the modelling of C-14 in the biosphere in the subsequent 30 years or so [e.g. EPRI, 1999; Avila and Pröhl, 2008].

A summary of the work that BIOPROTA has undertaken in relation to C-14 since 2005 was then given, which has comprised:

☐ literature reviews of the behaviour of C-14 in both terrestrial and aquatic ecosystems [Limer et al., 2011; Thorne et al., 2018];

☐ reviews of models, both conceptual and mathematical, used to represent C-14 in both terrestrial and aquatic ecosystems [BIOPROTA, 2005];

☐ application of models to both hypothetical and real-world scenarios of release of C-14 to terrestrial ecosystems [Limer et al., 2009, 2011, 2017; Mobbs et al., 2013; Thorne et al., 2018]; and

☐ a series of workshops to discuss the above [BIOPROTA 2010, 2013, 2014; Smith, 2015; Smith and Smith, 2014; Limer, 2017].

The work undertaken has sought to further our understanding as to the dynamics of the carbon pools in the systems of interest, to then be able to ascertain which parts of the environment are in equilibrium with each other and thus be able to assess the applicability of a specific activity model. Although the timeframes of interest for radioactive waste disposal are long (typically 10s or 100s thousands of years), the data available for model validation exercises typically addresses shorter-term fluctuations.

2.1.1 Terrestrial ecosystems

One area of interest with respect to terrestrial ecosystems is the chemical form in which the C-14 would enter the biosphere, either in gaseous or dissolved form. Of particular interest has been the potential for any C-14 bearing methane to be oxidised in the soil zone, with the resulting C-14 labelled carbon dioxide being available for plant uptake. Recent experiments [Shaw et al., 2018a,b] support the hypothesis of near-complete oxidation within a few tens of centimetre of unsaturated soil. This is something which has been reproduced reasonably well in modelling studies undertaken through BIOPROTA [Limer et al., 2017] and in studies undertaken outside of the group [Ota and Tanaka, 2019; see also Section 2.4.3 of this report].

With respect to plant uptake of carbon from the atmosphere, the work of BIOPROTA using hypothetical C-14 release scenarios indicated that was it was understanding the dynamics within the atmosphere that would be key to our being able represent such systems in our models [e.g. Limer et al., 2011]. The work highlighted that there were potentially significant conservatisms in the representation of diffusion in the canopy atmosphere, and gas exchange within that atmosphere and with the above-canopy atmosphere, in terms of the resulting calculated concentrations of C-14 in plants. Subsequent studies undertaken by BIOPROTA that have utilised real-world data sets have demonstrated that representation of the atmosphere that the plants obtains C-14 from via photosynthesis is indeed key [Limer et al., 2017; Thorne et al., 2018]. Again, these results are reflected in studies undertaken outside of the group [e.g. Aulagnier et al., 2012, 2013; Limer et al., 2015].

Uncertainties remain in the wider scientific community regarding the potential role of roots in the uptake of carbon by plants, with calculations undertaken by participants in the BIOPROTA C-14 studies and by other organisations indicating their potential importance (e.g. see Section 2.4.3 of this report).

2.1.2 Aquatic ecosystems

At the 2013 workshop, the dynamics of C-14 in aquatic environments were discussed, starting with a review of its behaviour and modelling approaches, plus overviews of data sets relating to C-14 in Duke Swamp, Canada, and C-14 in the North East Irish Sea arising from Sellafield discharges. Since then, a review of carbon in freshwater lakes and uptake into fish has been conducted through BIOPROTA (Section 2.2.1 of this report; see also Thorne et al. [2018]). The possible activities that BIOPROTA could undertake to further our understanding of the appropriate representation of carbon, and C-14, in aquatic systems for the purpose of safety assessment calculations were discussed further later in the workshop.

2.1.3 Continuing issues

Given the integral role of carbon in the environment, an understanding of the dynamics of the carbon pools in the systems of interest can achieved with site characterisation studies that are adequate to understand the spatial and temporal equilibrium or disequilibrium of those carbon pools, and of relevance to the assessment objectives. This then supports the arguments for the use of specific activity models versus models based on a mechanistic understanding of the system dynamics in representing C-14 dynamics in the biosphere for that system.

Finally, it was noted that the ICRP are currently re-evaluating C-14 dose coefficients, considering the chemical form of the carbon.

2.2 C-14 IN AQUATIC ENVIRONMENTS

2.2.1 BIOPROTA review of carbon in freshwater lakes and uptake into fish

Karen Smith (BIOPROTA C-14 project Technical Support Team) presented.

The uptake of C-14 from water by fish has been shown to be a dominating exposure pathway for both humans and non-human biota. If equilibrium is assumed between fish and water then high concentration ratios fish are derived, as fish have a higher stable carbon content than the water. However, the reality will depend on where fish obtain their carbon from. Determining this requires a understanding of the carbon dynamics in the system, including imports and exports of carbon and the characteristic timescales of turnover of different carbon pools.

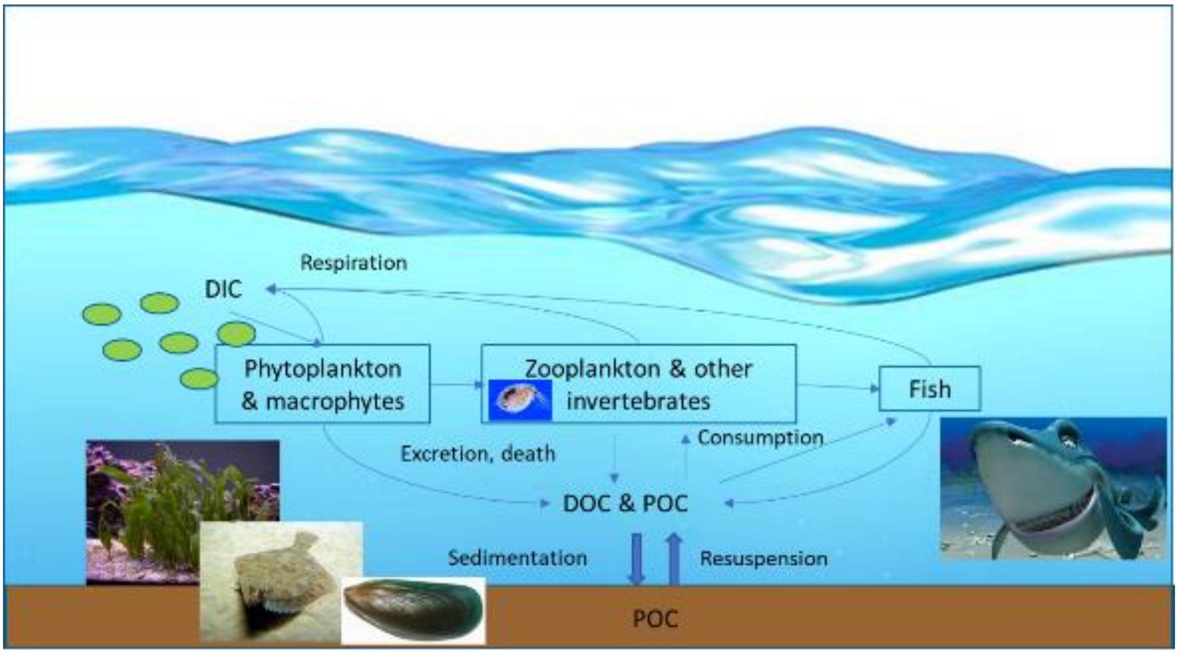

Figure 1: Freshwater lake food web

Part of the 2017/18 BIOPROTA project on C-14 included review of carbon cycling in freshwater lakes (see Figure 1), and is reported in Thorne et al. [2018]. The freshwater lake food web comprises phytoplankton, macrophytes, benthic microalgae, zooplankton and other invertebrates, and fish. All biota will respire, thus cycling carbon back to dissolved inorganic carbon (DIC). Excretion from and the death of biota will lead to the production of dissolved organic carbon (DOC) and particulate organic carbon (POC). Particularly for POC, there is then scope for sedimentation and resuspension.

There is a growing body of evidence that the terrestrial environment subsidises carbon demands of aquatic food webs, as aquatic systems degrade rapidly compared with terrestrial systems. Thus, terrestrial-POC (TPOC) and terrestrial-DOC (TDOC) enter the aquatic system, with field studies finding that terrestrial carbon is evident at all trophic levels in aquatic food webs. For example, some studies show 20-50% of zooplankton biomass is attributable to terrestrial carbon [Pace et al., 2004]. Up to 55% of the POC can come from terrestrial environment, and up to 100% of the DOC can come from the terrestrial environment [Wilkinson et al., 2013]. The contributions from terrestrial environments tend to be low for highly productive lakes. Conversely, some aquatic carbon can enter terrestrial systems, e.g. through consumption of fish by terrestrial biota.

There are many forms of carbon in terms of availability/degradation. Aquatic carbon tends to be labile, energetically rich and rapidly utilised. Terrestrial carbon typically has both labile and recalcitrant components. The labile terrestrial carbon enters the pelagic food web, whereas recalcitrant carbon enters the lake sediments and benthic food web. Both season and climate will affect the balance of terrestrial carbon entering the aquatic environment.

The review in Thorne et al. [2018] noted that methane is increasingly being recognised as a source of carbon into aquatic food webs [Jones and Grey, 2011]. Substantial amounts of methane can be produced in anoxic environments, which is then converted by methane-oxidising bacteria (MOB) at the anoxic-oxic interface in the sediments. The productivity of MOB can be high, with evidence of their consumption contributing to both the benthic and pelagic aspects of the freshwater lake food web [e.g.

Deines et al., 2007, 2009; Jones and Grey, 2011; Sanseverino et al., 2012]. A high TPOC input to the lake may provide a carbon source for methane production.

With respect to the carbon uptake by fish, there is evidence of the trophic transfer of both terrestrial carbon and methane throughout the lake food web. Some studies suggest that up to 50% of fish biomass comes from terrestrial carbon [Weidel et al., 2008]. Studies using stable C-13 have helped to identify the methane contribution to fish diets [e.g. Deines et al., 2009; Keaveney et al., 2015], and have shown the action of MOBs at oxic-anoxic interfaces has implications for C-14 methane entering lake food webs [e.g. Harrod and Grey, 2006].

The implications for safety assessments are that:

☐ terrestrial carbon can contribute a significant fraction (ca. 50%) of the carbon in an aquatic food web, and may therefore warrant explicit consideration in assessment calculations; and

☐ the action of MOB at oxic-anoxic interfaces has implications for C-14 labelled methane entering freshwater lakes and associated food webs.

Following the completion of the review, the following potential next steps were identified:

☐ extend the review to encompass other aquatic systems (e.g. rivers, marine);

☐ further review of carbon uptake routes (food versus direct uptake from water);

☐ consider various studies that are available detailing the transport of stable carbon isotopes through food webs; and/or

☐ review stable isotope studies to see if there are sufficient, coherent data to support a model scenario around carbon pools and fluxes in an aquatic system.

In the discussion following the presentation, it was noted that both SKB and Posiva have studied carbon balances as part of their site characterisation studies. For example, for Forsmark lakes, the conclusion was that the terrestrial carbon contribution was much less than 50%, and more like approximately 10%. It was also queried whether, outside of the nuclear field, studies into the terrestrial inputs of carbon to aquatic systems has/is being undertaken, and whether there are established modelling approaches that could support C-14 assessments.

2.2.2 Analysis of C-14 in marine biota samples

Maria Nordén (SSM) presented.

A Swedish study in 2006 measured the background C-14 level in macroalgae (244 Bq kgC^{-1}) and found that marine samples in the vicinity of the Barsebäck nuclear power plant (NPP, a boiling water reactor, BWR) were up to 15% higher than background levels [Stenström et al., 2006]. The same study noted that higher C-14 levels would be expected in the marine environment around pressurised water reactors (PWRs) than around BWRs.

A new project, combining researchers in the operational and waste disposal teams at SSM, started in 2015. The aims of this project are to increase the knowledge of the dynamics of C-14 following releases from NPPs into aquatic environments. The project started with a literature review and the validation of a field monitoring methodology with a pilot study in the vicinity of the Ringhals NPP (BWR). In particular, C-14 is being measured in seawater, bladder wrack (*Fucus vesiculosus*), blue mussel (*Mytilus edulis*) and fish (*Ctenolabrus rupestris, Symphodus melops*).

Following the pilot study, a sampling study between August 2018 and July 2019 is underway, again around Ringhals NPP. It is hoped that the outcomes of this study will be presented at the ICRER conference in 2020[a], and that the findings will also be published in an SSM report.

New methods for analysing C-14 in marina biota and in seawater have been developed. The method used has been relatively successful for marine biota, with around 80% chemical recovery of the C-14. However, there are issues around the availability of fish in the study area and also the recovery of C-14 in the seawater. A new strategy is being implemented regarding understanding the C-14 concentrations in seawater, where a literature review of the study of C-14 in seawater will be used, the reason for the low chemical recovery in the water will be investigated, and the current extraction method will be modified to improve the chemical recovery.

In the discussion following the presentation, interest was expressed in finding out details of the extraction method being used for seawater, with a view to supporting SSM in determining how the method might be driving the low chemical recovery rates. It was noted that it could be interesting to look at the differences between the reactors in terms of C-14 concentrations in seawater and marine biota, to get some information to see if such variability around nuclear facilities might be expected in countries other than Sweden.

It was noted that climate change may have implications on the behaviour of marina biota types (e.g. fish) in the longer term, though the data being collected as part of this study are focussed upon short-term releases, not radioactive waste disposal safety assessments.

2.2.3 Development of a dynamic model of C-14 in a freshwater stream

Kathryn Higley (Oregon State University, OSU) presented.

An assessment of a waste disposal site in the US has shown C-14 to be the dominant radionuclide in terms of doses to humans, due to exposure via the fish ingestion pathway. That analysis was based upon a specific activity model applied within the Hanford Environmental Dosimetry System (Generation II, or GENII[b]), with a chronic release scenario and reasonably maximally exposed individual (RMEI) pathway analysis. It was queried whether the use of a dynamic model and probabilistic analysis would give the same doses as the specific activity model, and whether such a model would permit the analysis of acute release events.

Prof. Higley has implemented a provisional dynamic C-14 model of the freshwater stream using the STELLA programming language platform, developed by ISEE Systems[c]. The probabilistic model input parameters for dose calculations were generated using Oracle Crystal Ball[d].

The conceptual and mathematical model developed for this study has utilised the Studsvik 'B' model from a BIOMOVS II study relating to lakes [BIOMOVS II, 1996] and papers by Steve Sheppard [e.g. Sheppard et al., 2006a,b]. The model includes inflow /outflow of constituents bearing carbon, variable influx of DIC and DOC/POC, DIC input from atmosphere to stream, and temperature effects on photosynthesis and respiration. Further developments of the model are planned, including the addition

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[a] https://www.icrer.org/

[b] https://energyenvironment.pnnl.gov/resources/resource_description.asp?id=93&type=tech

[c] https://www.iseesystems.com/store/products/

[d] https://www.oracle.com/applications/crystalball/

of hydraulic geometry relationships to support the representation of variations in streamflow, and the effects of light levels on photosynthesis and respiration.

The carbon model has six compartment (carbon dioxide dissolved in air, carbon dioxide dissolved in freshwater, carbon in fish, carbon in phytoplankton, DOC/POC and solid phase carbonate in surface sediments). Carbon and C-14 dynamics are modelled separately (Figure 2).

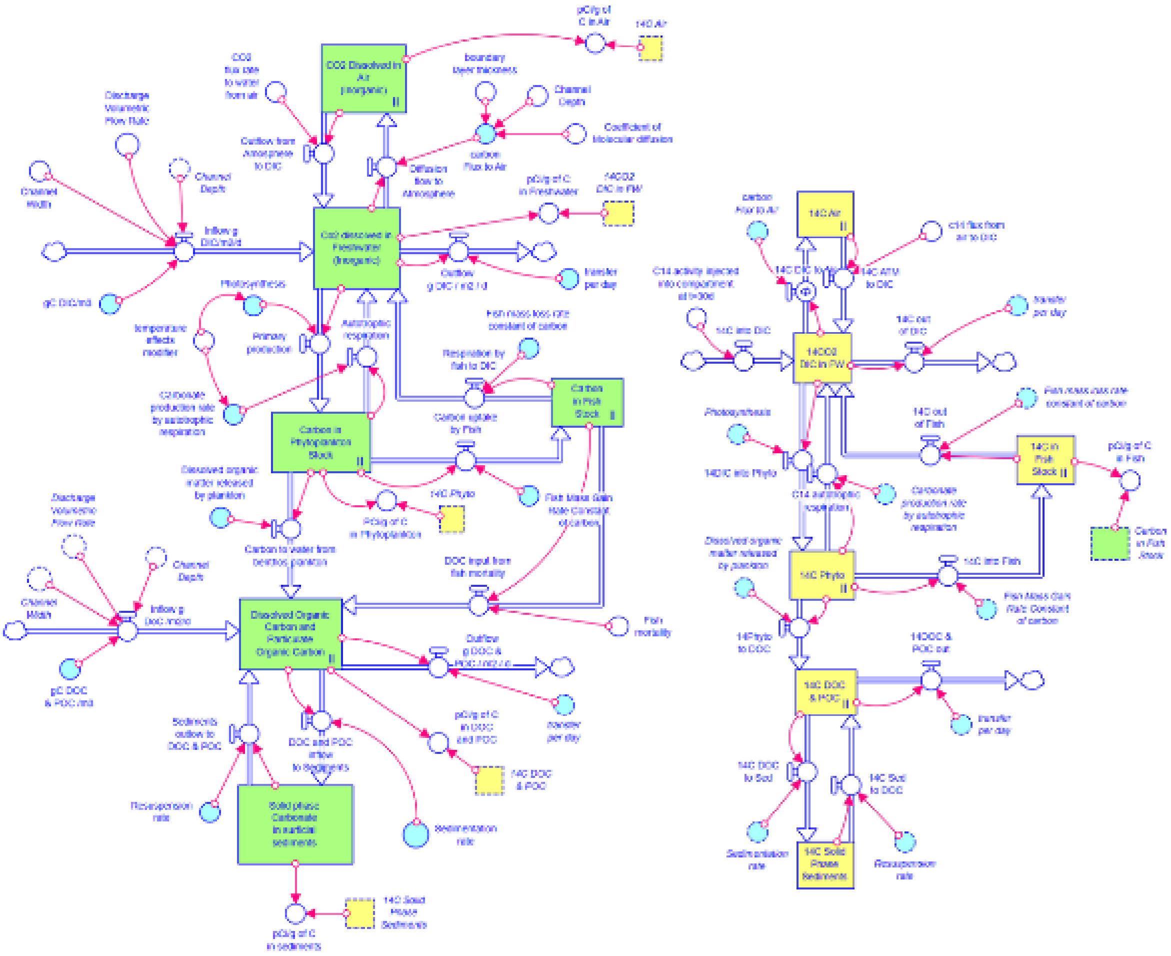

Figure 2: Carbon and C-14 models of a freshwater stream

The model includes several processes to represent the transport or conversion of carbon from one compartment to the other, some of which can be modified by abiotic factors relating to the system (e.g. temperature). Preliminary simulations have been run for a two-year period, using daily temperature variation, with an annual cycle, and a single pulse of C-14 into the stream at an elevated flow rate (15 times higher than usual). The Euler integration method has been used to calculate the activity concentration at each time step as a ratio of C-14 (pCi m^{-2}) and stable C (g m^{-2}) in each compartment.

Preliminary results were presented at the workshop, showing annual oscillations in the stable carbon stocks in the phytoplankton and fish, as expected due to temperature effects on phytoplankton photosynthesis, and accumulations in the stable carbon stocks in sediments and as dissolved carbon dioxide in air. The increase of C-14 in the phytoplankton reduces very quickly after the pulse input, with a longer retention time in the fish.

The calculated C-14 concentrations in fish were exported into Microsoft Excel, where probabilistic dose calculations were undertaken. The mass of fish consumed and associated distributions were derived from the U.S. Environmental Protection Agency (EPA) exposure factors handbook [EPA, 2011].

The hydraulic geometry calculations, which will be based on Leopold and Maddock [1953], will give hydraulic conductivity, flow rates, etc., based upon the geometry (width and depth) of the stream under consideration. The U.S. Geological Survey has historical monitoring data from streams that give temperature, pH etc. profiles of the water bodies being monitored.

Prof. Higley emphasised the provisional nature of the preliminary results, noting that the transfer rates are not in balance, as might have been expected. There is potential to investigate the implications of daily variations in streamflow rates (e.g. due to rainfall, runoff, etc.) would be on the transfer rates between compartments.

In the discussion following the presentation, the sensitivity of the calculated doses to the assumed human consumption rates and calculated concentrations of C-14 in fish was raised as a potential issue. It was queried whether the number of meals assumed in the calculations is a true reflection of the reality of diets in the vicinity of the site.

The nature of the of the surface water system being considered was also discussed, noting that there might be less scope for accumulation and recycling of C-14 in sediments in rivers and/or streams than there might be in the lake model on which the initial analysis is based. Furthermore, the streamflow rates may mean that the ‘phytoplankton’ pool would be replaced by benthic algae in driving the stream food web. In addition, it might be conceivable that the stream was flowing too quickly to sustain a population of fish at the density assumed.

Prof. Higley offered to circulate her mathematical model, so that other organisations could implement the model in their own preferred software codes.

2.3 C-14 FROM SUB-SURFACE

2.3.1 CAST Project: CArbon-14 Source Term: main outcomes and lessons learned

Maryna Surkova (FANC) presented.

There are many challenges associated with understanding in the potential release mechanisms of C-14 from radioactive waste materials. The data to support our understanding of C-14 release rates and speciation are sparse. For example, it has long been assumed that the C-14 released from irradiated metals is in a single chemical form and is released at a constant rate dependent on the corrosion rate and C-14 inventory. These estimates of long-term release rates from metals are based on the proportion of C-14 in the metal of interest, supported by short-term leaching experiment results, which may be more reflective of the labile carbon in the material.

The CAST Project[e] (CArbon-14 Source Term) was an EU project with 33 partner organisations, funded by the European Union's Euratom Seventh Framework Programme FP7/2007-2013 under the grant agreement No 604779. The scope of the project was to increase understanding in the potential release mechanisms of C-14 from radioactive waste materials, considering: wasteform dissolution and C-14 leaching, waste package corrosion, and the long-term evolution of the disposal system. By increasing understanding of these potential release mechanisms, the high-level objectives of the project were to:

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[e] https://www.projectcast.eu/

- decrease uncertainties in the long-term safety of radioactive waste disposal; and
- increase confidence in the safety case.

The project had work packages investigating the following four C-14 source terms: steels, Zircaloy, ion-exchange resins (IEX) and graphite. For each source term, work was undertaken to better characterise the source term with respect to inventory, release rates and speciation from those materials. In particular, the details objectives were to:

- improve scientific understanding of the C-14 release mechanisms and rates from:
  - the corrosion of irradiated steels and Zircaloys; and
  - the leaching of ion-exchange resins and irradiated graphite under geological disposal conditions; and
- provide chemical speciation of the C-14 released and how this relates to aqueous conditions.

With respect to steels, the objectives were to develop analytical techniques for identification and quantification of C-14 species formed during corrosion of irradiated steels under conditions relevant to cement-based repositories. The steels work package team also sought to validate existing activation models by measuring C-14 inventories in irradiated steel and carry out experiments and modelling to improve the understanding of the speciation and release rate of C-12/C-13 and C-14 from corrosion of steels. The main outcomes from this work package were that the C-14 inventory can be calculated from the nitrogen content of the steel and other metallic minerals together with the neutron flux during reactor operations. The inventory associated with the neutron flux is dependent on the position of the component(s) of interest in the reactor. However, there are often uncertainties associated with the actual amount of nitrogen in the steel, which can differ significantly from the amount reported by the manufacturer, and the exact position of the component in the reactor. This can lead to discrepancies of up to 300-400% between the modelled and measured C-14 inventories. Furthermore, there is currently no information on the fate and speciation of C-14 released under disposal conditions.

With respect to Zircaloy, the objectives were to obtain a better understanding of C-14 behaviour in Zr fuel claddings and speciation of released C-14. The work to support this involved an assessment of C-14 inventories in zirconium alloy metals and oxides, characterisation of the C-14 release from Zr corrosion and Zr oxide dissolution, and determination of the C-14 speciation under simulated disposal conditions. In contrast to the situation with steels, there was found to be good agreement between the modelled and measured C-14 inventory in Zircaloy. It was noted that the type of Zircaloy influences the total C-14 inventory and its speciation. The C-14 release is mainly dominated by the oxide. With respect to C-14 speciation, the organic fraction is more important than the inorganic, with the speciation evolving with time. Several methods were used to investigate the corrosion rates of Zircaloy, which found that irradiated Zircaloy generally corroded more rapidly than unirradiated Zircaloy. However, the underlying processes are still not fully understood, with more work being undertaken.

With respect to ion-exchange resins, the objectives were to obtain a better understanding of the C-14 source from spent ion-exchange resins from BWR and PWR. This was achieved by reviewing the current status and undertaking work to characterise the C-14 inventory and its speciation. Spent ion-exchange resins are almost always mixed together in storage tanks at the nuclear facility. Their ionic load and activity depend on: chemical or radiochemical saturation, maximum service lifetime, unusual pressure drop behaviour (clogging); and the fluid quality (becoming insufficient). The potential disposal of spent ion-exchange resins in a subsurface repository in a cementitious environment raises concerns regarding the potential for uncontrolled releases of C-14 from the spent ion-exchange resins.

There is scarce information regarding speciation of C-14 accumulated in spent ion-exchange resins. The working group found some unexpected partitioning of the C-14 between inorganic (carbonate) and organic fractions (predominantly small molecular weight molecules such as formate, acetate and oxalate), with the majority of the C-14 activity arising from the inorganic part. However, there are no published data regarding the leaching of organic molecules from spent ion-exchange resins, and there are no reliable isotopic or scaling factors for C-14 available for spent ion-exchange resins. Any proposed scaling factors should be regarded with scepticism as their basis upon a mixture of types of spent ion-exchange resins, their multiple possible origins and their diverse ageing and degradation processes under varying service and storage conditions means that any proposed values would be highly questionable.

With respect to graphite, the objectives were to understand the factors determining the release of C-14 from irradiated graphite. This was achieved by determining the C-14 inventory in graphite and its spatial distribution, measuring the release rate and speciation of C-14 to solution and gas in contact with aqueous solutions, and determination of the impact of selected waste treatment options on C-14 release. The working group found that a substantial fraction of the C-14 in graphite will not be releasable. That which can be released will have some quick and some slower release mechanisms, with location in the graphite having the potential to affect the release rates (Figure 3).

In the discussion following the presentation, it was agreed that whilst scaling factors can be used for some of the materials, this cannot be done for spent ion-exchange resins, as the spent ion-exchange resins will be mixed. The release routes should be clearly identified for each material type. For Posiva, spent ion-exchange resins are one of the most important sources of C-14.

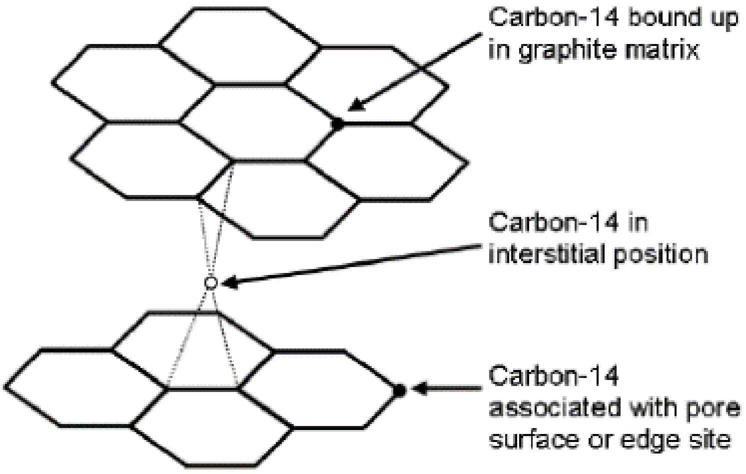

Figure 3: C-14 potential locations in irradiated graphite [Swift et al., 2016]

2.3.2 Recent C-14 assessment developments in Finland

Pekka Kupiainen (Posiva) presented.

Since the TURVA-2012 assessment was submitted, there have been some C-14 assessment updates at Posiva. These include use of the S-VAT model developed for SKB [Avila and Kovalets, 2016], some new data for assessment parameters, and also updated inventory calculations.

Posiva are using a specific activity model, with a more detailed model structure and data uncertainty treatment, based on the S-VAT model structure for relevant biotopes (forest, mire, croplands, coastal and inland aquatic biotopes). The model has been implemented in the Ecolego software[f]. Assuming that the system reaches equilibrium rapidly, the S-VAT model has been used to develop equivalent “transformation factors” for the C-14/C-stable ratio calculated for unit releases. Posiva also undertook an analysis of data uncertainties.

Posiva has undertaken some modelling of a forested mire, calculating concentration profiles and the effects on dose assessment. The new model has higher calculated concentrations of C-14 as compared with the Avila and Pröhl (2008) model. Conversely, the new model calculates lower concentrations of C-14 in trees than the Avila and Pröhl (2008) model. The results so far are for a unit source term. The new model calculates understorey C-14 concentrations in vegetation four orders of magnitude greater than those of trees. There will be follow-on work to use the TURVA-2012 source term.

Consideration of C-14 assessment in the safety case context was presented. Validity of the specific activity model(s) have improved, but there are remaining questions around the time resolution used in the validation as compared with the validation data, spatial resolution, and model simplifications versus system understanding.

The C-14 assessment is part of the “assessment chain” and reflects focus on key parts/radionuclides in the system. More extensive datasets that could further help with validation (representativeness issues) and balance in comparison with other parts of the safety case should be considered. The models should be validated for a range of source terms. For example, the Natural Resources Institute Finland (www.luke/fi/en) has collected some data, which Posiva have been analysing.

In the discussion following the presentation, it was noted that Posiva intend to update their aquatic model for C-14 to account for losses to the atmosphere. The possibility of linking the aquatic and terrestrial C-14 systems was also discussed, to keep the C-14 balanced in the overall biosphere. The benefits of adding such complexity to the assessment level model balanced against the importance of the pathways and effect on calculated doses was discussed. It was suggested that, if it were possible to show that the loss of C-14 from a water body to the atmosphere is trivially small, then it would be viable to argue that there was no need to couple the atmospheric parts of the terrestrial and aquatic ecosystems.

2.3.3 Improved C-14 modelling in SE-SFL & work in progress

Peter Saetre (SKB) presented.

The C-14 model for the SE-SFL assessment is an evolution of the model used in SR-PSU [Saetre et al., 2013]. As previously, the conceptual model is based on understanding of stable carbon cycling, although stable carbon dynamics are not routinely calculated in parallel as part of the assessment. The following primary conceptual and mathematical evolutions of the modelling were presented:

- ☐ continuous cultivation of farmland;
- ☐ further analysis of gas exchange and diffusion modelling in different environments; and
- ☐ a review of methane dynamics.

The conceptual model for gas exchange in lakes assumes that there is no significant coupling between the water body and the atmosphere above, such that it is appropriate to model the effect of degassing

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[f] http://ecolego.facilia.se/ecolego/show/HomePage

and gas uptake as gross transfers, with the gas exchange derived on that basis. Models developed by Cole and Caraco [1998] and MacIntyre et al. [2010] for the water-atmosphere boundary layer have been used to support this aspect of the modelling.

For the gas exchange in saturated soils found in mires, the modelling is based on Wania et al. [2010]. The SR-PSU assessment [SKB, 2015] found that the assumed piston velocity parameter had to be calibrated downward (to one-third of its initially assumed value) to yield realistic DIC concentrations in the oxic region of the soil. This reduction is consistent with observations from lake ecosystems, where emissions of carbon dioxide and methane can be reduced by up to 60% if a mat of floating aquatic vegetation is present [Attermeyer et al., 2016]. In the SE-SFL assessment, the peat depth model and DIC concentrations are run as a stable C model in a “ghost” mode as part of the assessment, to help validate the C-14 model.

Fick’s second law has been used to model gas diffusion in unsaturated soils, with the Millington and Quirk [1961] model used to represent the dependency of the diffusion coefficient on soil properties. One of the biggest controls on the modelling of gas diffusion in unsaturated soils is the assumed air-filled porosity, with a higher porosity generally leading to more rapid diffusion; having a “ghost” stable carbon model of this aspect of the system will help support the assumed degassing rates used in the C-14 assessment. Results from sensitivity calculations were presented, where it was shown that the turnover rate in soil had little effect on the calculated activity concentrations in the soil and atmosphere. However, it was noted that root uptake of C-14 had the potential to make calculated doses from a terrestrial system larger than calculated doses from the consumption of fish.

SKB has commissioned a review of sources, transport and losses of methane in the biosphere. The review has found regional estimates of methane fluxes from wet environments, but there are no site-specific estimates and there is only limited knowledge of methane production and oxidation in sediments. SKB has, therefore, initiated field studies on methane coming from sediments and from groundwater. They are collecting data at a number of sites and also from some specific streams. It was argued that the lack of knowledge justifies the simplifying (and cautious) assumption of complete methane oxidation in mires and aquatic ecosystems. In upland soils, the characteristic length of full methane oxidation is a few tens of centimetres. If the water table is a few times below this level, then methane oxidation would be nearly complete in those soils. Such arguments are supported by the findings of the RWM-funded experiments at the University of Nottingham [Atkinson et al., 2014; Shaw et al., 2018a,b]. Assumptions regarding methane production in mires and aquatic ecosystems is felt not to be critical for the safety assessment, as drainage and cultivation of soils results in similar, or higher, calculated doses than releases to mires or aquatic ecosystems.

Attention was brought to a paper reporting a field study to investigate the spatiotemporal variability of lake pCO_2 and CO_2 fluxes in a hemi-boreal catchment [Natchimuthu et al., 2017]. Their monitoring studies have found evidence of variability in pCO_2 in three lakes, and terrestrial carbon inputs into the lakes are were also observed.

In the discussion following the presentation, it was clarified that the C-14 model used in the SE-SFL assessment will be compared to the previous C-14 assessment models, in terms of outputs, as part of the SE-SFL assessment. This is both to build confidence in SKB’s modelling internally and also to demonstrate responsiveness to the regulator.

The return of carbon to the soil was discussed. It was noted that whilst the top layer of the soil is of fixed depth in the model (30 cm), the post-glacial sediments grow during lake development, and peats might grow on land. Root decay also provides a mechanism for carbon to re-enter the soil. Whilst some might argue that C-14 transfers via such processes are so diluted as to not warrant consideration of its recycling, SKB like to have mineralisation and consideration of peat growth etc. explicitly included in their assessment models.

2.3.4 C-14 model for application to assessment of HLW disposal in Germany

Alex Diener (BfS) presented.

The administrative regulation in Germany is currently being developed to calculate the exposure of the representative person from a HLW repository. BfE (the Federal Office for the Safety of Nuclear Waste Management) will specify the approach for considering radionuclide transport in the host rock, and BfS (the Federal Office for Radiation Protection) will develop statutory provisions relating to the biosphere. As two separate offices are preparing the regulations, there is a need to be clear as to assumptions at the geosphere-biosphere interface zone, which for gaseous transport is the boundary between solid rocks and soil or receiving waters, and for water-mediated transport in groundwater bodies which migrate into surface waters or that are located close to the surface and could be used for irrigation, livestock watering and drinking water for humans.

C-14 is one of the radionuclides that BfS is focussing on because it is one of the radionuclides that produces the highest calculated dose contributions and carbon (and C-14) has its own characteristic cycle in the biosphere. The conceptual and mathematical biosphere model that BfS is developing for C-14 assessments was presented. The model used for C-14 is relatively simple, covering the basic underlying processes, with the doses providing an indication as to the risks associated with HLW, not predictions of real doses.

For soil/groundwater transport into lakes and rivers, a constant rate of gaseous C-14 is assumed to transport from the host rock directly into receiving waters. That rate will depend upon the activity concentration and the host rock type, needing detailed modelling to support assessment modelling assumptions. BfS plans to use 3D modelling to calculate advective groundwater flow and groundwater transport within the rock/soil matrix. Activity concentrations in stagnant water bodies, such as lakes and swamps, are currently being modelled using the “small lakes” model that applies to lakes (less than or equal to 400 km^2) from the IAEA SRS-19 [IAEA, 2001]. For rivers, consideration is given to the radionuclide concentration along the flow path, accounting for input into the river, groundwater runoff, the mixing ratio between groundwater and river runoff and the travel time of water through the river.

For terrestrial ecosystems, exposure pathways include ingestion of crops and grazing cattle. The gaseous transport from host rock goes directly into the soil, and then directly into the canopy atmosphere. A constant release rate is assumed, dependent upon the activity concentration of C-14, the chemical form (CO_2 or CH_4) and the soil matrix. The amount of C-14 entering the soil via irrigation is time dependent, varying according to differences between precipitation and evapotranspiration. A specific activity model is used to calculate the concentration of C-14 in plants from $^{14}CO_2$ in the atmosphere, distinguishing between plants used for grazing and those plants grown for direct human consumption. A slightly different approach is used to model the uptake of C-14 in plants due to irrigation with contaminated water. A standard transfer factor is used for contamination of animal products.

Humans can be exposed via ingestion and inhalation, and six age groups are considered by BfS. Dependent upon the scenario considered, exposure may come from a combination of pathways, with habits based on present-day data. The inhalation dose accounts for the chemical speciation of the C-14.

In the discussion following the presentation, the use of six age groups was queried. Alex noted that BfS had found differences in the calculated doses for infants if consuming breast milk or formula, so have decided to stay with six age groups, rather than three, or even one, used by other organisations. The variety in terms of regulatory guidance with respect to exposure groups, including age groups and activity groupings is being considered further by the collaborative IAEA MODARIA WG6/BIOPROTA work to review and enhance the BIOMASS methodology.

2.3.5 BIOPROTA Duke Swamp study

Russell Walke (BIOPROTA C-14 project Technical Support Team) presented.

Duke Swamp is on the edge of the Canadian Nuclear Laboratoies (CNL[g]) Chalk River research site, by the Ottawa River, Canada. Historical disposals of C-14 of ~100,000 m^3 low level radioactive waste (LLW) between 1963-2006 in Waste Management Area A (WMA-C), with an engineered cover installed in 2013 (on the basis of historical leachate monitoring). There have been studies over the years to understand the geology and hydrology of the site [e.g. Donders et al., 1996]. The leaching of C-14 has been known about for some time. Within the swamp area, sampling by CNL looked at soil/sediment and sphagnum moss and the spatial distribution of C-14 within these media [e.g. Milton et al., 1998; Yankovich et al., 2014]. More recently, Jon Napier (an OSU PhD student) looked at C-14 concentrations between WMA-C and the swamp [Napier, 2015].

There is considerable uncertainty regarding the exact inventory of C-14 disposed in WMA-C. Cautious estimates of 4000 GBq have been considered, with trial pits and groundwater sampling indicating 3 to 6 GBq/y reach the swamp in groundwater [Killey et al., 1998]. Estimates of C-14 in the swamp range from 7.6 to 24.8 GBq, with some estimates made of losses due to volatilisation in the swamp.

The Duke Swamp data was first considered within a BIOPROTA project on C-14 via a modelling scenario in 2016, commencing with a collation of the available information. Model development and calculations were undertaken in 2017-2018, with the aim to broadly reproduce observed concentrations in groundwater, sediment and vegetation. The Technical Support Team undertook some calculations in AMBER[h], as documented in the associated report [Thorne et al., 2018].

At first a single layer (with respect to soil depth) model was considered, with a spatial extent away from the source (Figure 4). Dissolved C-14 and fixed C-14 were explicitly represented. Initially, the compartment length assumed was too large (50 m), with numerical dispersion dominating the results. Refining the compartment length to 0.2 m improved the modelling, with the system reaching equilibrium within a few hundred years. However, in a single layer model the volatilisation losses were too high, and so the calculated C-14 concentrations did not penetrate very far into the swamp.

The model was therefore extended to have two-layers of soil (Figure 5). The upper soil layer was 0.3 m deep and the lower soil layer 2.7 m deep. Rather than using a uniform compartment length, the compartment length increased from 1 m to 10 m with distance from the groundwater source term as the model progressed into the swamp. As with the first iteration of modelling, carbon fixed in organic matter and that available in soil water were explicitly represented in each layer (Figure 6), with the fraction of carbon volatilising from each layer differing.

These changes led to improved representation of the C-14 dynamics in the swamp, which are discussed in more detail in Thorne et al. [2018]. The results, using the 6.5 GBq/y source term, reflect the observations fairly well (Figure 7). The analysis implies that the behaviour in organic soils can vary greatly, due to spatial distinctions in organic matter turnover rates and in the degree of water saturation. At Duke Swamp, ~20% of carbon released from organic matter by mineralisation is likely to be reincorporated in new biomass. This is a considerably larger fraction than would be expected for

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[g] CNL = Canadian Nuclear Laboratory, formerly part of AECL

[h] https://www.quintessa.org/software/AMBER

agricultural crops growing on mineral soils, and probably relates to degree of soil aeration and canopy structure.

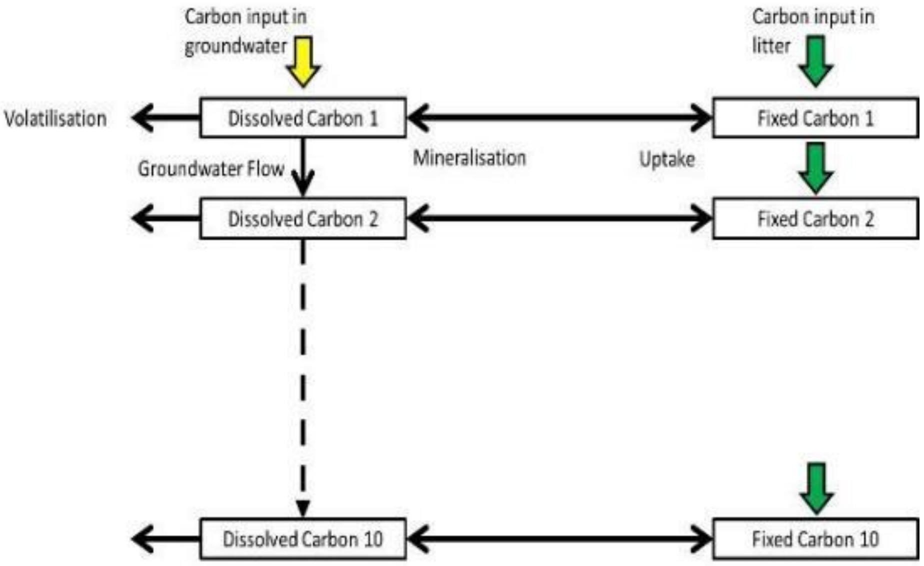

Figure 4: Single soil layer 1D model of Duke Swamp

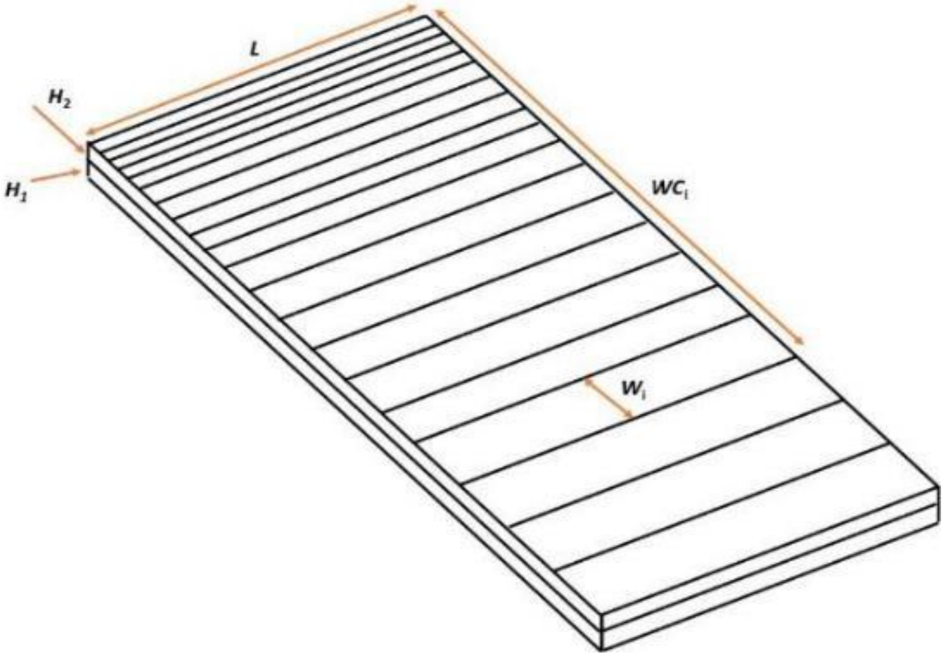

Figure 5: Two-layer geometry of Duke Swamp model

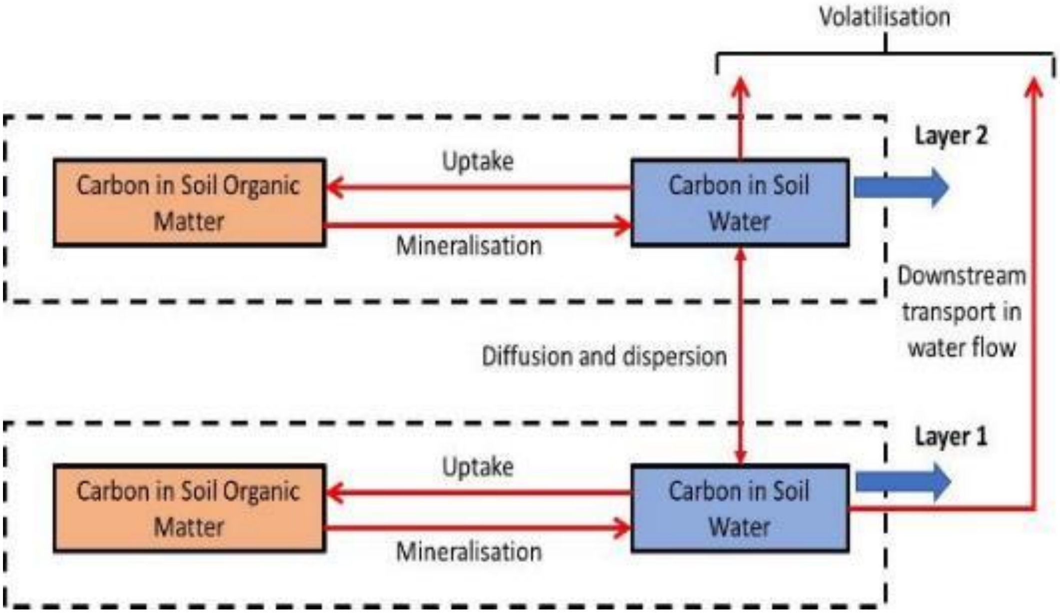

Figure 6: Two-layer Duke Swamp model structure

In the discussion following the presentation, it was recognised that whilst some of the model parameterisation came from site measurements and observations [e.g. Killey et al., 1998; Yankovich et al., 2014], there are other parts of the parameter space that might fit the monitoring data as well. There was some discussion of different modelling approaches and around the definition of processes and parameters. The key is having processes that are understood and can be parameterised, and then explain the role of the equations and parameters. It was noted that such work could help identify key pathways, parameters (degree of saturation, organic matter content).

It was suggested that, in light of the results of the work presented, other organisations might be interested in modelling Duke Swamp[i]. It was queried whether there might be other existing sites, that might be similar and with monitoring data available, that could be used to build further confidence. One suggestion was the US Savannah River Site [e.g. Cummins et al., 1991; Savannah River Nuclear Solutions, LLC, 2018].

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[i] Peter Saetre (SKB), expressed an interest to be sent the input data.

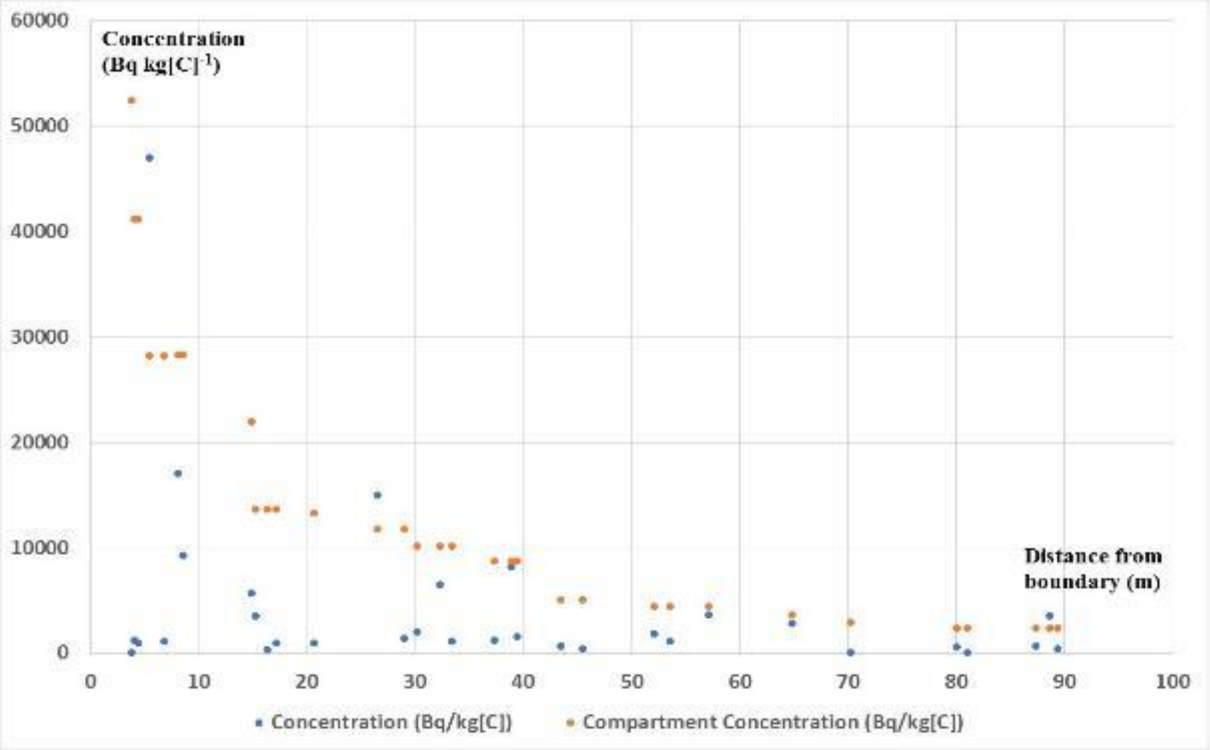

Figure 7: C-14 in soil water as a function of distance from the entry point into Duke Swamp (Bq kgC^{-1}); measured concentrations of C-14 are shown in blue, with modelled concentrations in orange

2.4 C-14 IN THE ATMOSPHERE

2.4.1 BIOPROTA study on C-14 deposition to pasture at La Hague

Russell Walke (BIOPROTA C-14 project Technical Support Team) presented.

The Orano La Hague nuclear fuel reprocessing plant in Northwest France has been operating since the late 1960s and has authorised atmospheric discharges of approximately 15 TBq y^{-1}. Between September 2006 and July 2008, IRSN monitored C-14 at a site 2 km north of the La Hague site. Data relating to meteorological conditions, source terms, atmospheric concentrations, soil and pasture concentrations were collected. Hourly measurements of Kr-85 in the atmosphere at the monitoring site, combined with monthly measurements of Kr-85 and C-14 in the atmosphere, were used to derive an implied hourly C-14 atmospheric source term at the site. Considerable variation in the C-14 concentration in the atmosphere at the monitoring site was observed (Figure 8).

The field data collected by IRSN provided an opportunity to validate the modelling of C-14 uptake into pasture from the atmosphere. Therefore, as part of a BIOPROTA study, five organisations[j] applied their models to the La Hague scenario [Limer et al., 2017]. The models varied in structure and processes explicitly represented.

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[j] Andra, FANC, IRSN, SSM and SKB.

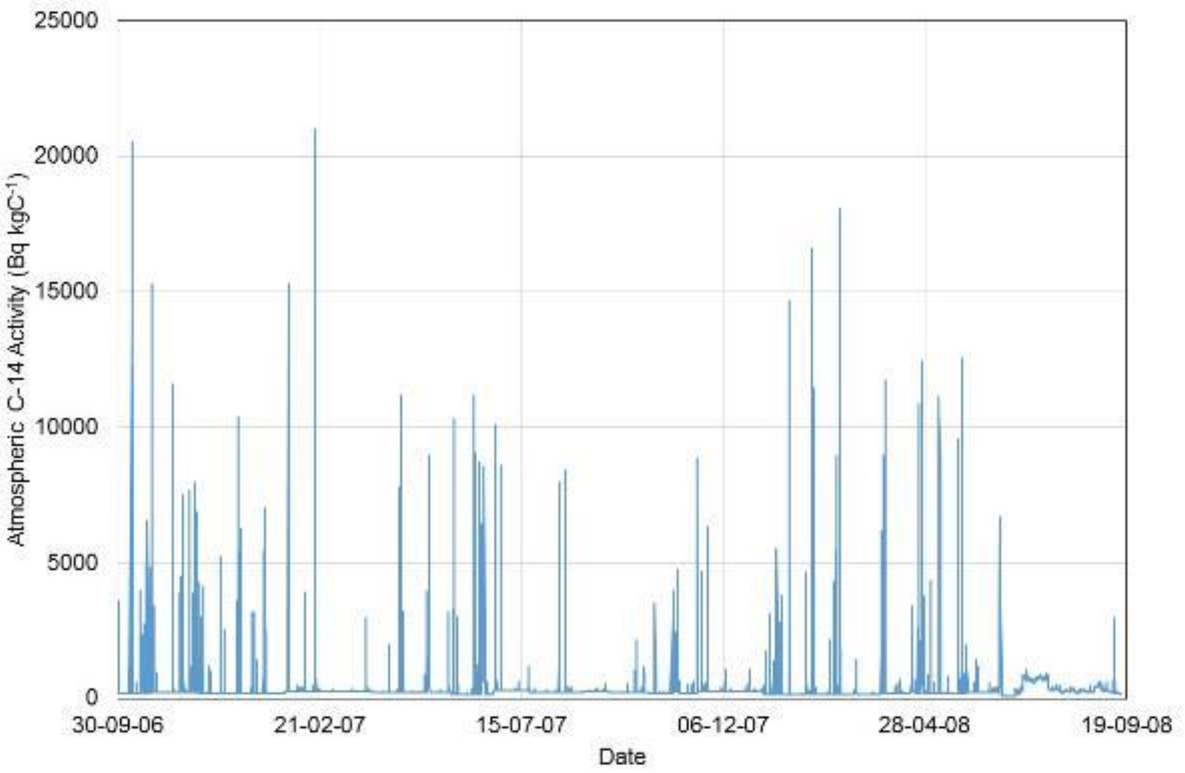

Figure 8: High temporal resolution C-14 atmospheric concentrations (Bq kgC^{-1})

The models used the same atmospheric source term, and aimed to calculate C-14 concentrations that followed the observed C-14 concentrations in the pasture. The models were adapted to accommodate the high-temporal resolution of input data and performed well when compared against the observational data (Figure 9).

The calculated soil concentrations of C-14 were more variable, with a dependency on the assumed initial conditions. However, for an atmospheric source term the dominance of that means that the soil compartment(s) do not significantly affect the key results of crop concentrations.

The results of this study build confidence in the modelling of C-14 dynamics in plants exposed to an atmospheric source term.

In discussion, it was noted that with the well-defined pasture growth information and the atmospheric source term, it would be viable to calculate plant C-14 activity without dynamic modelling. Recycling of carbon, and C-14, back into the soil could benefit from further characterisation.

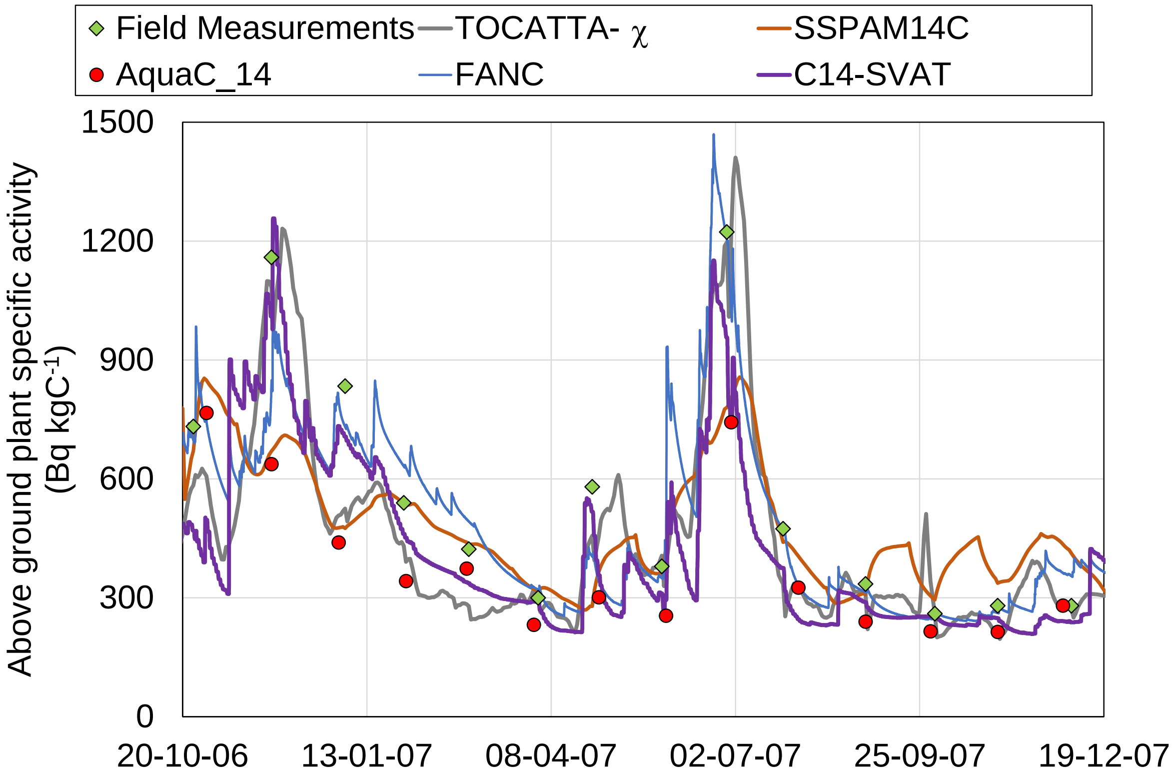

Figure 9: Calculated C-14 concentrations in pasture (Bq kgC^{-1}) for the La Hague scenario (reproduced from Limer et al., 2017)

2.4.2 Simplifying TOCATTA-χ: development of a meta-model for predicting C-14 (and H-3) activity concentrations in agricultural ecosystems

Séverine Le Dizès-Maurel (IRSN) presented.

TOCATTA is a dynamic model of transfer of C-14 and H-3 from the air to soil-plant systems, which is used by EDF to carry out impact studies within the SYMBIOSE tool. It operates on a daily time-step, using time-dependent plant growth curves and meteorological data. It is assumed that there is isotopic equilibrium between the plant compartment and the canopy atmosphere at each time step of the calculations, with the results mainly driven by the daily meteorological conditions and atmospheric $^{14}CO_2$ concentration.

To verify the conceptual model, IRSN undertook the ‘VATO’ project, in which monitoring at the Orano La Hague nuclear fuel reprocessing plant was used to demonstrate high temporal variation in the atmospheric concentrations of C-14 within a single day, leading to substantial variations in plant C-14 concentrations being observed. Recognising that the daily time-step of TOCATTA was not adequate to predict plant activity concentration in response to short-term acute variations of the atmospheric source term ($^{14}CO_2$), such as had been seen at La Hague [Aulagnier et al., 2012], a mechanistic process-based model was developed. This model, TOCATTA-χ, is better able to predict the response in plant C-14 concentrations to continuous or acute releases of C-14 to the atmosphere [Aulagnier et al., 2013]. However, the data requirements for such a model are high.

IRSN has been working on the implementation of an aggregated TOCATTA model[k], which will ultimately be implemented in SYMBIOSE in 2020. It is parameterised for grass, working on a daily timestep. It is a combination of the TOCATTA and TOCATTA-χ models, in both conceptual and mathematical aspects. The plant types and path growth rates are as TOCATTA-χ, with the canopy and soil models from TOCATTA. There is a new canopy photosynthesis model, derived from a leaf photosynthesis calculation of the Farquhar model used in TOCATTA-χ [Farquhar and von Caemmerer,1982].

As a starting point, the TOCATTA-χ model calculates several variables (e.g. evapotranspiration, photosynthesis, stomal conductance and respiration) and the calculates plant C-14 concentrations on an hourly basis. A basic sensitivity analysis (using Tornado plots) is then used to derive an aggregated and simple set of equations on the basis of the most sensitive variables of TOCATTA-χ with respect to the calculated plant C-14 concentration. The analysis at this stage identified eight key parameters: leaf area index (LAI), foliar nitrogen content, mean daily temperature, daily temperature range, solar radiation, day of the year, latitude, and difference of soil water potential.

The aggregated equations based on these eight variables were then calibrated in order to produce similar estimations of photosynthesis (and stomatal conductance), as the ones obtained by TOCATTA-χ. This was achieved using the Monte-Carlo method with Latin Hypercube sampling, and approximately 500 calculation runs. With these equations, hourly dynamics of the system, including C-14 concentration in plants, could be calculated based upon daily inputs.

The aggregated TOCATTA model was then applied to the Orano La Hague dataset, to understand whether or not significant bias might have been introduced into the model in the process of aggregating TOCATTA and TOCATTA-χ. An initial comparison between the three TOCATTA models, and also SSPAM^{14}C developed by SSM [Limer et al., 2015], against the measured grass C-14 activity was presented (Figure 10).

IRSN will be preparing a paper for a peer-reviewed journal discussing the development of the aggregated TOCATTA model, and the comparisons with the other TOCATTA models, and SSPAM^{14}C. Other organisations were invited to supply calculated C-14 activity in plants up to the end of December 2008. The input information used for the previous BIOPROTA project [Limer et al., 2017] would be made available to support those calculations.

In the discussion following the presentation, the contribution of C-14 contained in rain to the plant C-14 concentration was queried. As the contribution from C-14 in rain is negligible, although there is a rain model within TOCATTA, it is ignored in terms of the potential for any C-14 in the rain to contribute to plant or soil C-14 concentrations. The term “meta” in the presentation title was used to reflect the intermediate level of complexity of the aggregated TOCATTA model. It was noted that much of the model overhead is associated with the calculation of plant growth rates etc. It was suggested that the modelled biomass could be validated against the measured biomass growth.

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[k] The mathematical model is implemented in GoldSim.

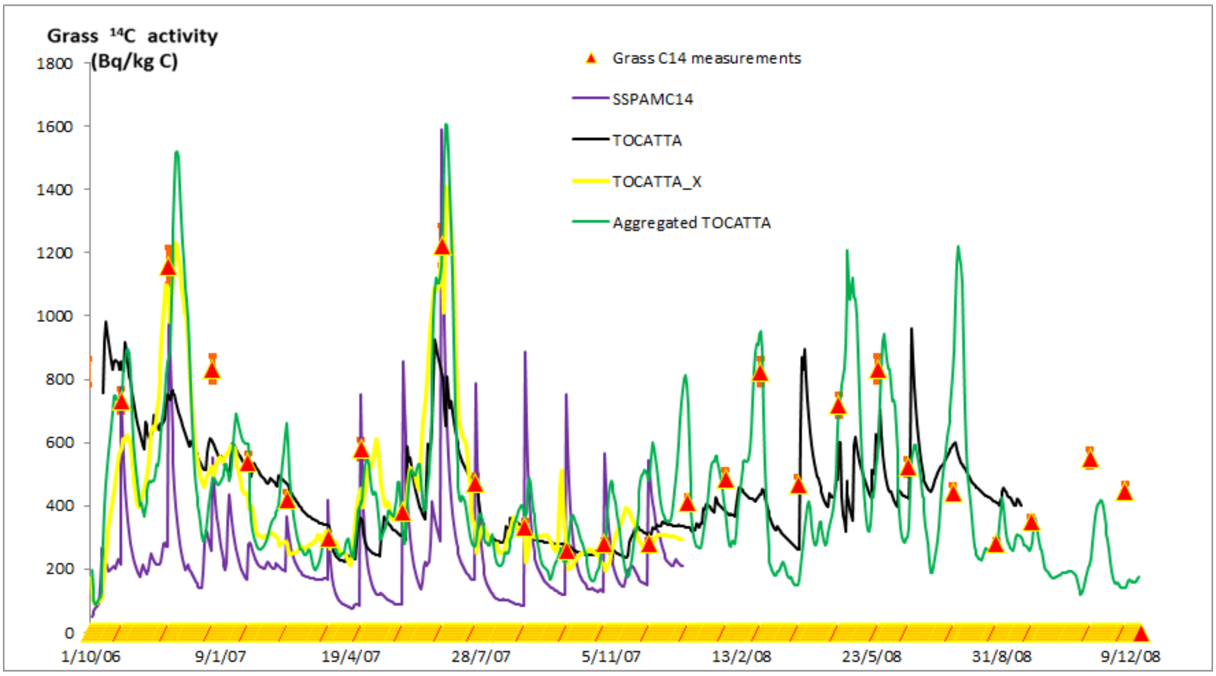

Figure 10: Initial comparison of calculated plant C-14 activity by the TOCATTA models, and also SSPAM^{14}C, against the measurements taken in the vicinity of the Orano La Hague nuclear fuel reprocessing plant

2.4.3 Introduction of the JAEA's C-14 model, SOLVEG-II

Masakazu Ota (JAEA) presented.

The presentation was a summary of a paper that has recently been published in the Journal of Environmental Radioactivity [Ota and Tanaka, 2019].

It is important to be able to predict the transfer of C-14 from soil to vegetation. The key uptake pathways are foliar uptake (stomatal deposition) and root uptake. If the source is to the atmosphere, then foliar uptake is expected to be much greater than soil uptake, and even if the source is from the soil foliar uptake is expected to be greater than root uptake. Most models focus on the foliar uptake, with some neglecting the root uptake. From a geological disposal facility, one might expect the C-14 leaving the saturated zone to be in the form of methane, and then require oxidation to carbon dioxide before entering the plant, either via root or foliar uptake. Interest was in the extent of root uptake influencing C-14 transfer to plants during $^{14}CH_4$ release from a GDF.

JAEA sought to investigate how and to what extent does the root uptake of $^{14}CO_2$ influence the C-14 transfer to plants during $^{14}CH_4$ release from a deep disposal facility. In particular, they sought to test the hypothesis that the deep distributions of roots and methane oxidation in a soil would result in a greater root-uptake of $^{14}CO_2$ than foliar uptake of $^{14}CO_2$. To investigate this, JAEA developed a 1D land surface $^{14}CH_4$ transport model, SOLVEG-II. The model has compartments for $^{14}CO_2$ in soil air, soil water, atmosphere, leaf cellular water and C-14 in leaf organic matter. It has compartments to represent $^{14}CH_4$ in soil air and the atmosphere. The model allows methane oxidation in all soil layers, but no methane oxidation in the aboveground atmosphere.

SOLVEG-II was applied to the University of Nottingham dataset. The results show that the observed rapid decrease in $^{14}CH_4$ levels in the soil layers, both temporally and spatially, is best reproduced by the calculations if both diffusion and oxidation are included in the mathematical model. For the $^{14}CO_2$, the model predicted increased and decreased activity levels as seen in the field data. The calculated fluxes from the ground surface are similar to those measured in the experiments, within a factor of 2-3. Using these model outputs, the concentrations of C-14 in the soil and atmosphere required to determine the ratio of root and foliar uptake of C-14 could be calculated.

Consideration was then given to the extent to which C-14 root uptake occurs in plants. A hypothetical $^{14}CH_4$ source term at the bottom of a 1 m thick soil, with differing rooting zone assumptions (shallow, deep) and methane oxidation profiles with soil depth (shallow, intermediate, deep) were considered. These sensitivity studies illustrate that for the system considered:

- when shallow roots and shallow oxidation were assumed, the calculated root uptake of C-14 into the plant was less significant than foliar uptake (20% from roots);
- when shallow roots with intermediate and deeper oxidation were assumed, the calculated root uptake contribution increased a little (22% and 21% respectively);
- when deep roots were assumed, the calculated root contribution to the uptake of C-14 in the plant increased substantially, from 63% for shallow oxidation, up to 100% for intermediate and deep oxidation.

In the discussion following the presentation, it was noted that the University of Nottingham dataset used related to vegetated soil measurements taken in the field. Whilst other datasets could be used for application with this model, it was not immediately apparent if other suitable datasets might exist. With respect to the root profiles, clarifications were made regarding the maximum depth of the roots. For the shallow roots, the maximum depth was assumed to be 11 cm below the surface. For the deep roots, the maximum rooting depth assumed was 97 cm, a reflection of the presence of trees. It was noted that roots tend to be denser closer to the soil surface, and that it is not enough to consider only the root profile, but it is also where the plants are getting their water from. A need to validate the root profile and water uptake assumptions was recognised.

Particular concern was raised as to the source of bulk carbon assumed to be used to generate new plant biomass. It was noted that comparisons could be made to the modelling of Cl-36 and radioactive caesium in soil-plant-atmosphere systems, and the interplay with the stable element dynamics.

2.4.4 Model of Soil to Vegetation to Atmosphere Transport of C-14 (SVAT-C14)

Rodolfo Avila (Facilia/AF Consult) presented.

The SVAT-C14 model was developed to consider continuous releases of C-14 as carbon dioxide from soil over an area covered by grass, crops and forests. Inputs with irrigation water are included in the model when appropriate for the vegetation being considered. The model calculates concentrations of C-14 in plants and in air above the release surface, the latter being used for the calculation of inhalation doses.

The earliest models developed for SKB assumed that all the C-14 released from a GDF would enter the plant directly, which was a highly conservative of assumption. The conceptual model evolved to consider an atmosphere with mixing above some fixed height [Avila and Prohl, 2008]. With this model, it was shown the calculated plant C-14 concentrations were dependent on the assumed mixing height, which is non-trivial to quantify. To further investigate the sensitivity of the calculated C-14 concentration in plants to atmospheric conditions, a process-based 1D Lagrangian transport model has been developed, SVAT-C14 [see Figure 11; Avila and Kovalets, 2016; Kovalets et al., 2018]. The model was validated against a dataset from the Norunda field station [Kovalets et al., 2018], and also data from the SMEAR-II dataset collected in Finland. The calculated C-14 concentrations in plants were in reasonable agreement with datasets collected at monitoring stations in the forest, but did not perform so well for the station at the edge of the forest. It is felt this is because the model considers only a 1D environment, and so does not account for the potential for lower vegetation to be present in front of the tree layers.

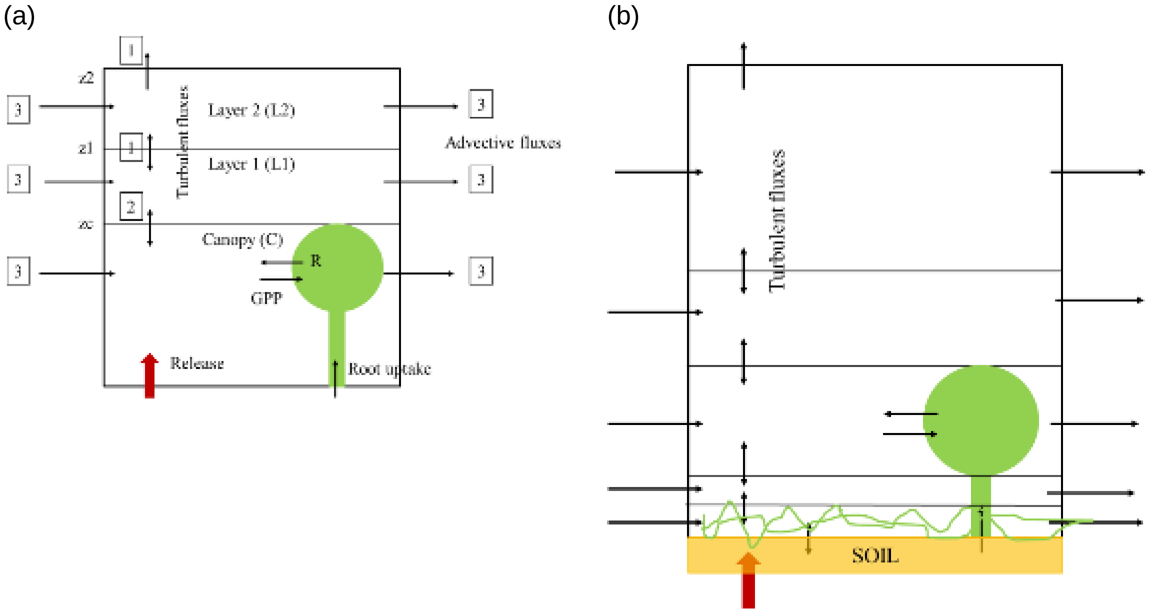

Figure 11: Schematic of SVAT-C14 model. (a) crops and grasses; (b) trees.

Further analysis has been carried out, considering different types of forest (e.g. rock forest, heath forest, grove forest, and forested mire), and agricultural crops, such as barley. Consideration has been given to the sensitivity of the calculated C-14 concentrations in vegetation to input parameters such as vegetation properties, CO_2 concentrations, meteorological conditions and area of release, both with and without advective transport of C-14 in the atmosphere in the modelling. This has shown a clear dependency of the calculated plant C-14 activity on assumptions regarding advective transport in the atmosphere and also the area of release of C-14 bearing gas to the aboveground atmosphere. The work reported in Avila and Kovalets [2016], is in the process of being updated to incorporate the findings of these sensitivity studies.

In the discussion following the presentation, it was noted that the impact of the assumed fraction of uptake by roots upon the calculated plant C-14 activity is very small in the sensitivity analysis.

2.4.5 BIOPROTA Finnish forest study

Russell Walke (BIOPROTA C-14 project Technical Support Team) presented.

The University of Helsinki has been monitoring material and energy flows at four forest locations in Finland for over 20 years, studying atmosphere-vegetation-soil interactions. These monitoring stations, each known as a "Station for Measuring Ecosystem-Atmosphere Relations" (SMEAR), provide data sets for research understanding.

The SMEAR II station in Hyytiälä, Southern Finland was established in 1995. It is dominated by 57-year old (in 2019) Scots Pine trees, with homogeneous vegetation for around 200 m in all directions around the flux towers. The key measurements being collected are the atmospheric $\Delta^{13}C$ and $\Delta^{14}C$ values at four different heights above the ground surface, where Δ represents the difference of the activity in the sample concentration in comparison to a reference 1950 atmospheric concentration. The data collected shows a clear seasonal variation in the $\Delta^{13}C$ and $\Delta^{14}C$ measurements at all four heights, but that there is little evidence of the isotopic ratio of C-13/C-14 changing with height above ground level, or distance below/above the canopy. Furthermore, there is relatively little variation in $\Delta^{13}C$ and $\Delta^{14}C$ soil atmosphere values with soil depth.

A detailed 1D physically based model was developed for the site, with the outcomes of the model application reported in Kovalets et al. [2018]. A simpler, analytical, modelling approach was adopted by the BIOPROTA Technical Support Team; the results are presented in Thorne et al. [2018].

The atmospheric C-14 concentrations calculated using the detailed model attenuate very quickly above the soil surface, consistent with the field observations, though there are very few data points for direct comparison. Similarly, the simple analytical model can also be used to explain the observed attenuation of the flux of C-14 gas from the soil. The rate of uptake of canopy atmosphere carbon (and thus C-14) and the diffusive and dispersive mixing rates are shown to be key controls on the C-14 activity in the canopy atmosphere.

3. DISCUSSION AND RECOMMENDATIONS

3.1 IDENTIFICATION OF GAPS IN UNDERSTANDING THAT NEED TO BE ADDRESSED IN BUILDING FURTHER CONFIDENCE IN ASSESSING C-14 IN THE BIOSPHERE

3.1.1 C-14 in aquatic ecosystems: developments and issues remaining to address

At this workshop, discharges of C-14 into the marine environment, rivers and freshwater lakes were all covered in the presentations. Aquatic systems are still an important aspect of C-14 dose assessment modelling, both for radioactive waste disposal and operational discharges. Certainly, there are opportunities for further work to validate modelling assumptions and build further confidence.

One aspect of the behaviour of aquatic systems that has not been well studied with respect to radioactive waste disposal safety assessment is the input of terrestrial carbon into aquatic systems, and the potential for there to be some two-way exchange of carbon between terrestrial and aquatic systems. There is potential to review models currently used in safety assessments for radioactivity in the environment, as well as models of stable carbon in terrestrial-aquatic systems. The review could identify interactions between terrestrial and aquatic systems that are explicitly considered in those models, and what data are available to support, or refute, those assumptions.

A conceptual model for DIC, DOC and POC in water bodies, and the interactions with the terrestrial environment, could be developed within BIOPROTA to provide a generic structure for rivers, marine systems, freshwater lakes etc. that other organisations might use as part of their safety assessment calculations.

It was recognised that mechanistic models are very important in allowing simple models to be justified. A scenario like the Duke Swamp area is very interesting – it is a good analogue for surface repositories, has a good set of data and different models could be used to test against observations – which would allow consideration of the different pathways and whether they need to be explicitly represented.

It was noted that IRSN and EdF are currently undertaking some modelling with SYMBIOSE, supported by field monitoring (see below), of the overall behaviour of carbon in a river system.

Multiple participants expressed an interest in undertaking a model-model, ideally model-model-data, comparison study for a freshwater system, either a river or a lake. It was noted that a river water model would be extremely valuable for operational and disposal groups and that it would be good to bring together those groups to address the issue.

Several potential datasets that could be available to support the validation of C-14 freshwater models were discussed.

- Llyn Trawsfynydd, United Kingdom. This is a man-made lake next to the former Trawsfynydd nuclear power plant. There is monitoring data going back to the 1960s of discharges of radioactivity in the lake and activities measured in rainbow trout, the latter including C-14 activities.

- Shield lakes area, Canada. This involves a shield lake system that was subject to C-14 additions in the 1970s, specifically into Lakes 224 and 226, and monitored thereafter for approximately 15-20 years [e.g. BIOMOVS II, 1996; Bower, 1981; Hesslein et al., 1980; Stephenson et al., 1994, 1995]. Samples of water, sediment, plants, invertebrates and fish were taken, though not all aspects of the systems were monitored concurrently.

- Swedish lakes. It was noted that SKB has published information on the carbon balance in several lakes that have been part of their field studies, which could provide more data to support the modelling of such systems, particularly on DIC, DOC and POC concentrations.

- ☐ River Rhine, France. EdF and IRSN are undertaking monitoring of radionuclide and chemical dispersion in the river, which includes C-14 measurements. Queries will be made regarding the potential for BIOPROTA to access this monitoring data, to support the validation of river models.

- ☐ Rivers and tributaries downstream of French NPPs. In autumn 2019, a PhD student will start at IRSN, acquiring some field data in rivers and tributaries to look at speciation of C-14 downstream of a NPP release. They will be looking at C-14 in a natural environment, measuring DIC, DOC, POC and COF (organic carbon in the dilution phase). COF is depleted in C-14, and tributaries with a lot of COF may have any industrial inputs of C-14 diluted significantly. There is also CIF, which is the inorganic form. There is no modelling anticipated as part of this PhD, but biogeochemical modelling with the data collected by other organisations would be welcomed.

It was agreed that the BIOPROTA Technical Support Team would review what data are available and make suggestions for potential next steps. One step would be a definition of what work could be undertaken to identify key uncertainties that still remain. Potential collaborative research topics should be defined in terms of helping to address those uncertainties.

Looking forward to other international programmes, it was noted that there might be an opportunity under the IAEA MODARIA III programme to undertake a project to review regulatory decisions that have been made relating to C-14 and focus resources on those problems that were identified.

3.1.2 Sub-surface and atmospheric C-14 releases to terrestrial ecosystems: developments and issues remaining to address

Prior to 2017, the work of BIOPROTA in this area has focussed upon agricultural ecosystems, with more recent work on both forest and wetland ecosystems. For many countries with temperate, boreal and/or arctic climatic conditions, these three broad ecosystem types cover the main potential terrestrial biosphere systems of interest.

It was noted that the Duke Swamp dataset could be useful for building further confidence in models of releases of C-14 from the sub-surface. In particular, it would be good to draw in additional models into the comparison. It was suggested that the Technical Support Team could refine the Duke Swamp scenario description to highlight the data available regarding hydrology etc., to help organisations (e.g. SKB, SSM) build a case internally for applying their models to such a system.

There still is an unanswered question regarding the extent of root uptake of carbon, and C-14, by plants. As has been demonstrated by the work presented in Ota and Tanaka [2019], even if a small percentage of total plant carbon is assumed to enter via the roots, if the C-14 specific activity in the soil atmosphere is much greater than that of the above-ground canopy atmosphere, then there is scope for the root uptake of C-14 to play an important role in dose assessment calculations. It was suggested that a review of the assumptions regarding plant uptake of total carbon from roots be revisited (e.g. is 2-15% still valid?), and that consideration be given as to the proportion of carbon (and C-14) from roots that is lost to the atmosphere rather than being incorporated into new plant biomass. The exchange of carbon, and C-14 in the canopy, via leaf gas exchange, was also noted as an area of uncertainty in our system understanding that could be addressed in future work.

As illustrated by the La Hague scenario, in some contexts, little consideration need be given to C-14 dynamics in soils in assessment modelling. Nonetheless, for sub-surface sources of C-14, soil dynamics may play an important role in determining potential consequences. It was suggested that it may be worthwhile to review the approaches used to model soil carbon (and C-14) dynamics in soil in operational and waste disposal models, and evaluate those approaches against the more recent literature on soil carbon cycling. For the context of radioactive waste disposal assessments, consideration needs to be given to the return of carbon (and C-14) to soils and implications for the persistence of C-14 in soil, and the associated residence time. The stable carbon cycle in soil could be reviewed and used to determine if radiological dose assessment models can be improved. The soil carbon cycle has linkages with the cycling of other elements relevant to radioactive waste disposal, such as Cl and Cs.

Over recent years, a number of waste management organisations (e.g. LLWR Limited, RWM, SKB) have developed new or refined models for C-14 in terrestrial ecosystems, with a conceptual model of the dynamics of carbon in the atmosphere based on micro-meteorological studies of canopy atmospheres of different types of vegetation. It was suggested that there could be scope for further comparison of such models, to look at the degree of dilution of C-14 from the sub-surface for differing vegetation canopies (e.g. crops, forests, mires). Posiva is going to be supporting an MSc thesis undertaking further analysis of the SMEAR monitoring data, and will try to ensure that BIOPROTA members can gain access to the thesis, once completed.

3.1.3 General discussion

The balance of complexities and degrees of conservatism in assessment models for operational discharges and models for assessment of radioactive waste disposal, were discussed. Understanding of the degree of conservatism in modelling assumptions is needed in each context. For radioactive waste disposal, the objective is typically a pessimistic comparison against regulatory limits, rather than a prediction. For operational assessments, there is more scope for realistic assumptions, given potential for validation through monitoring, though compliance assessments will remain conservative.

It is important to look at key parameters that could give rise to safety issues – model comparison output can be very helpful. The BIOPROTA studies help to explore the credibility of models proposed, whether these are in line with those used in other countries and also whether they reflect appropriate understanding of the modelled system.

BIOPROTA has previously undertaken work on the geosphere biosphere interface zone (GBIZ) that could be relevant to assessment of C-14. For example, that study encompassed the spatial areas of which a release might occur, and speciation in the GBIZ, both of which are of relevance to C-14.

As a general point, it was suggested that a future study could review the questions asked by regulators in review of licence/authorisation applications rather than simply presenting models. It was also noted that the use of academic process-based modelling to support simplification assumptions for our safety assessment modelling should be reflected in the updates to the BIOMASS guidance.

3.2 RECOMMENDATIONS

From the discussions held during the workshop, it is clear that there are areas in which additional collaborative work within the BIOPROTA forum could help to build further confidence in assessments of C-14 in the biosphere. In particular, the following potential activities arose as suggestions.

- Aquatic ecosystems:
    - Further review of interactions between terrestrial and aquatic carbon, for ecosystems other than lakes, e.g. rivers.
    - Development of generic conceptual model(s) for C-14 behaviour in freshwater systems (lakes and/or rivers), including uptake into the food chain.
    - Investigation and preparation of data set(s) that could be used for model-model-data comparison exercises in the future, including:

    - Llyn Trawsfynydd, United Kingdom;
    - Canadian shield lake;
    - Swedish lakes studied for SKB; and/or
    - River Rhine, France.

- ☐ Terrestrial ecosystems:
  - Facilitation of further model-model-data comparisons against the Duke Swamp data set.
  - Further investigation as to role of root uptake in overall plant carbon cycle and implications for C-14 uptake.
  - Review of soil modelling approaches.

These topics will be reviewed by the Technical Support Team, with a view to preparing and distributing proposals for further collaborative work to BIOPROTA members.

4. REFERENCES

Atkinson BS, Meredith W, Snape C, 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. Radioactive Waste Management Ltd.

Attermeyer K, Flury S, Jayakumar R, Fiener P, Steger K, Arya V, Wilken F, van Geldern R, Premke K (2016). Invasive floating macrophytes reduce greenhouse gas emissions from a small tropical lake. Nature Scientific Reports, 6:20424 | DOI: 10.1038/srep20424.

Aulagnier C, Le Dizès S, Maro D, Hébert D, Lardy R, Martin R, Gonze M-A (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 C, Le Dizès S, Maro D, Hébert D, Lardy R, Martin R (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.

Avila R, Kovalets I (2016). Models of C 14 in the atmosphere over vegetated land and above a surface-water body. SKB R-15-09, Svensk Kärnbränslehantering AB.

Avila, R., Pröhl, G., 2008. Models used in SFR 1 SAR-08 and KBS-3H safety assessments for calculation of 14C doses. Svensk Kärnbränslehantering AB (SKB) report R-08-16. Stockholm, Sweden.

BIOMOVS II (1996). Validation Test for Carbon-14 Migration and Accumulation in a Canadian Shield Lake. BIOMOVS II Technical Report No.14, September 1996.

BIOPROTA (2005). Model Review and Comparison for C-14 Dose Assessment, Theme 2 Task 3 Report. Main Contributors: S Sheppard (Task Leader) and M C Thorne.

BIOPROTA (2010). C-14 Long-Term Dose Assessment: Data Review, Scenario Development, and Model Comparison. Workshop Report, Version 2.0.

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. 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.

Bower PM (1981). Addition of Radiocarbon to the Mixed-Layers of Two Small Lakes: Primary Production, Gas Exchange, Sedimentation and Carbon Budget. PhD. Thesis Faculty of Pure Science, Columbia University, 236 pp.

Cole JJ, Caraco NF (1998). Atmospheric exchange of carbon dioxide in a low-wind oligotrophic lake measured by the addition of SF_6. Limnology and Oceanography 43: 647–656.

Cummins CL, Hetrick CS, Martin DK (1991). Radioactive Releases at the Savannah River Site 1954-1989 (U). Westinghouse Savannah River Company Report WSRC-RP-91-684.

Deines P, Bodelier PLE, Eller G (2007). Methane-derived carbon flows through methane-oxidizing bacteria to higher trophic levels in aquatic systems. Environmental Microbiology 9:1126-1134.

Deines P, Wooller MJ, Grey J (2009). Unravelling complexities in benthic food webs using a dual stable isotope (hydrogen and carbon) approach. Freshwater Biology 54: 2243-2251.

Donders RE, Killey RWD, Franklin KJ, Weich SJ, Strobel GS (1996). Trial Coring in LLRW Trenches at Chalk River, AECL report AECL-11681.

EPRI (1999). Carbon-14 in Low-Level Waste. Electric Power Research Institute report TR-107957.

Farquhar GD, von Caemmerer S (1982). Modeling of photosynthetic responses to environmental conditions. In: Lange, O.L., Nobel, P.S., Osmond, C.B., Ziegler, H. (Eds.), Physiological Plant Ecology II. Encyclopedia of Plant Physiology, New Series, vol. 12B. Springer Verlag, Heidelberg, pp. 549-587.

Harrod C, Grey J (2006). Isotopic variation complicates analysis of trophic relations within the fish community of Plußsee: a small, deep, stratifying lake. Archiv fur Hydrobiologie 167: 281-299.

Hesslein RH, Broecker WS, Quay PD, Schindler DW (1980). Whole-lake radiocarbon experiment in an oligotrophic lake at the Experimental Lakes Area, Northwestern Ontario. Canadian Journal of Fisheries and Aquatic Science, 37: 454-463.

IAEA (2001). Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment. Safety Report Series No.19. International Atomic Energy Agency, Vienna, Austria.

Jones RI, Grey J (2011). Biogenic methane in freshwater food webs. Freshwater Biology 56: 213-229.

Keaveney EM, Reimer PJ, Foy RH (2015). Young, old, and weathered carbon – part 2: Using radiocarbon and stable isotopes to identify terrestrial carbon support of the food web in an alkaline, humic lake. Radiocarbon Journal 57(3): 425-438.

Killey RWD, Rao RR, Eyvindson S (1998). Radiocarbon speciation and distribution in an aquifer plume and groundwater discharge area, Chalk River, Ontario. Applied Geochemistry, 13: 3-16.

Kovalets I, Avila R, Mölder M, Kovalets S, Lindroth A (2018). Verification of a One-Dimensional Model of CO_2 Atmospheric Transport Inside and Above a Forest Canopy Using Observations at the Norunda Research Station, Boundary Layer Meteorology, 168(1): 103-126.

Leopold LB, Maddock T (1953). The hydraulic geometry of stream channels and some physiographic implications, U.S. Geological Survey Professional Paper, 252, 57 pp.

Limer L (Ed.) (2017). C-14 Terrestrial Model-Data Comparisons. Report of an International Workshop. Version 1.0 (Final). www.bioprota.org.

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

Limer LMC, Smith K, Albrecht A, Marang L, Norris S, Smith GM, Thorne MC, 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.

Limer L, Les Dizès-Maurel S, Klos R, Maro D, 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.

Limer L, Ota M, Tanaka T, Thorne M, Walke R (2017). C-14 Terrestrial Model-Data Comparisons: Final Report. Report prepared within the BIOPROTA international collaborative forum QRS-1769A-1, Version 1.0, 11 July 2017. Available from www.bioprota.org.

MacIntyre S, Jonsson A, Jansson M, Aberg J, Turney DE, Miller SD (2010). Buoyancy flux, turbulence, and the gas transfer coefficient in a stratified lake. Geophys. Res. Lett. 37: 2–6.

Millington RJ, Quirk JP (1961). Permeability of porous solids. Transactions of the Faraday Society, 57: 1200-1207.

Milton GM, King KJ, Sutton J, Enright S (1998). Tracer studies of carbon source utilization in a wetland on the Canadian Shield. Journal of Applied Geochemistry 13: 23-30.

Mobbs S, Shaw G, Norris S, Marang L, Sumerling T, Albrecht A, Xu S, Thorne M, Limer L, Smith K, 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.

Napier JB (2015). Analysis of Carbon-14 Transport from a Contaminated Groundwater Source. PhD Thesis, Radiation Health Physics, Oregon State University.

Natchimuthu S, Sundgren I, Gåfalk M, Klemedtsson L, Bastviken D (2017). Spatiotemporal variability of lake pCO_2 and CO_2 fluxes in a hemiboreal catchment, Journal of Geophysical Research: Biogeosciences, 122: 30-49, doi:10.1002/2016JG003449.

Ota M, Tanaka T (2019). Importance of root uptake of $^{14}CO_2$ on ^{14}C transfer to plants impacted by below-ground $^{14}CH_4$ release. Journal of Environmental Radioactivity, 201: 5-18.

Pace ML, Cole JJ, Carpenter SR, Kitchell JF, Hodgson JR, Van de Bogert MC, Bade DL, Kritzberg ES, Bastviken D (2004). Whole-lake carbon-13 additions reveal terrestrial support of aquatic food webs. Nature 427: 240-243.

Sanseverino AM, Bastviken D, Sundh I, Pickova J, Enrich-Prast A (2012). Methane carbon supports aquatic food webs to the fish level. PLoS ONE 7(8): e42723. doi:10.1371/journal.pone.0042723.

Saetre P, Nordén S, Keesmann S, Ekström P-A (2013). The biosphere model for radionuclide transport and dose assessment in SR-PSU. SKB R-13-46, Svensk Kärnbränslehantering AB.

Savannah River Nuclear Solutions, LLC (2018). Savannah River Site: Environmental Report for 2017. SRNS-RP-2018-00470.

Shaw G, Atkinson BS, Meredith W, Snape C, Lever DA, Hoch AR (2018a). Methane Transport through an Agricultural Soil Following Pulse Injection of Isotopically-Enriched Methane in the Sub-surface. NERC Environmental Information Data Centre. https://doi.org/10.5285/18939865-d863-498b-b4cb-5661eaeadcfc.

Shaw G, Atkinson BS, Meredith W, Snape C, Lever DA, Hoch AR (2018b). Methane transport in agricultural soil after injection of isotopically-enriched methane in the sub-surface. Sci. Data 5, 180208. https://doi.org/10.1038/sdata.2018.208.

Sheppard SC, Ciffroy P, Siclet F, Damois C, Sheppard MI, Stephenson M (2006a). Conceptual approaches for the development of dynamic specific activity models of ^{14}C transfer from surface water to humans. Journal of Environmental Radioactivity 87: 32-51.

Sheppard SC, Sheppard MI, Siclet F (2006b). Parameterization of a dynamic specific activity model of ^{14}C transfer from surface water to humans. Journal of Environmental Radioactivity 87: 15-31.

SKB (2015). Handling of biosphere FEPs and recommendations for model development in SR-PSU, SKB R-14-02, Svensk Kärnbränslehantering AB.

Smith GM, Fearn HS, Smith KR, Davis JP, Klos R (1988). Assessment of the Radiological Impact of Disposal of Radioactive Waste at Drigg, NRPB-M148.

Smith K (Ed.) (2015). Long-term Dose Assessment for Carbon-14: Report of an International Workshop. Report prepared within the BIOPROTA international collaborative forum, Version 2.0, August 2015. Available from www.bioprota.org.

Smith K, Smith G (Eds) (2014). C-14: Data, Ecosystems and Dose Assessment: Report of an International Workshop, Aix-en-Provence, 1 – 3 April 2014. Report prepared within the BIOPROTA international collaborative forum. Available from www.bioprota.org.

Stenström K, Leide Svegborn S, Magnusson Å, Skog G, Zakaria M, Mattsson S (2006). Analysis of C-14 at nuclear facilities, industries and laboratories. SSI P 1378. Lund Internal Report LUNFD6/NFFR-3101.

Stephenson M, Motycka M, Schwartz W J (1994). Carbon-14 Activity in Water, Sediments and Biota of Lakes 226 North and 226 South and 224, Experimental Lakes Area, 1989 to 1994. AECL Research Technical Record, TR634 COG-94-97.

Stephenson M, Rowan DJ, Kelly C, Schwartz WJ, Motycka MF, Roshon RD (1995). Fate and Distribution in Sediments of Carbon-14 Added to Canadian Shield Lakes of Differing Trophic State. Limnology and Oceanography, 40: 779-790.

Swift BT, Swanton SW, Miller W and Towler G (2016). Carbon-14 Project Phase 2: Irradiated Graphite Wastes. Amec Foster Wheeler report for Radioactive Waste Management Limited AMEC/200047/004 Issue 2.

Thorne M, Smith K, Kovalets I, Avila R, Walke R (2018). C-14 in the Biosphere: Terrestrial Model-Data Comparisons and Review of Carbon Uptake by Fish. A BIOPROTA report.

U.S. Environmental Protection Agency (EPA) (2011). Exposure Factors Handbook: 2011 Edition. National Center for Environmental Assessment, Washington, DC; EPA/600/R-09/052F. Available from the National Technical Information Service, Springfield, VA, and online at http://www.epa.gov/ncea/efh.

Wania R, Ross I, Prenctice IC (2010). Implementation and evaluation of a new methane model within a dynamic global vegetation model: LPJ-WHyMe v1.3.1. Geoscientific Model Development, 3: 565-584.

Weidel B, Carpenter S, Cole J, Hodgson J, Kitchell J, Pace M, Solomon C (2008). Carbon sources supporting fish growth in a northern temperate lake. Aquatic Sciences 70: 446-458.

Wilkinson GM, Pace ML, Cole JJ (2013). Terrestrial dominance of organic matter in north temperate lakes. Global Biogeochemical Cycles 27: 1-9.

Yankovich TL, King-Sharp KJ, Carr J, Killey RWD, Beresford NA, Wood MD (2014). Spatial analysis of Carbon-14 dynamics in a wetland ecosystem (Duke Swamp, Chalk River Laboratories, Canada). Journal of Environmental Radioactivity, 137, 173-180.

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