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Swedish Radiation Safety Authority

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Modelling Approaches to C-14 in Soil-Plant Systems and in Aquatic Environments

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SSM perspective

Background

BIOPROTA (www.bioprota.org) is an international collaboration forum which seeks 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. Particular emphasis is placed on data required for the assessment of long-lived radionuclide migration and accumulation in the biosphere, and the associated radiological impact, following discharge to the environment or release from solid waste disposal facilities.

The project described in this report was supported financially by the following sponsoring organisations: Agence Nationale pour la Gestion des Déchets Radioactifs (Andra,France), Electricité de France (EDF, France), the National Cooperative for the Disposal of Radioactive Waste (Nagra, Switzerland), the Nuclear Decommissioning Authority –Radioactive Waste Management Directorate (NDA RWMD, UK), Nuclear Waste Management Organisation of Japan (NUMO, Japan), Nuclear Waste Management Organization (NWMO, Canada), Posiva Oy (Finland), Svensk Kärnbränslehantering AB (SKB, Sweden) and Strålsäkerhetsmyndigheten (SSM, Sweden).

The report presents the results of a range of reviews of field measurements, experimental work and model development and application concerning C-14 behaviour in the environment. It also describes presentations and discussions held during an international workshop on 12th to 14th February 2013, in Stockholm, hosted by SKB. The workshop provided further input and peer comment on work in progress at a wider variety of institutions in many countries.

Objectives

The objectives of the project were:

1. to review wider literature on C-14 in soil-plant systems;
2. to review recent model developments on C-14 in soil-plant systems;
3. to review key features of models and data for assessing doses to humans via consumption of freshwater biota following C-14 long-term release to surface water bodies;
4. to identify the key issues relating to modelling of terrestrial and aquatic pathways for C-14; and
5. to develop recommendations for future research activities to address these issues.

Results

Substantial expertise on carbon biogeochemistry already exists in the fields of plant physiology and aquatic ecology and therefore is it beneficial to draw on this to identify the important issues. A paper describing the BIOPROTA C-14 model inter-comparisons, and plans for the forward programme was published in Radiocarbon Journal. Further studies on C-14 (including modelling, model inter-comparisons, and experimental work) have been on-going by individual BIOPROTA member organisations and continued interest was shown in further collaborative work, including assessment of doses to humans due to long-term releases of C-14 to surface water bodies.

Noting the above, an international workshop was held from 12 - 14 February 2013, in Stockholm, to enable joint review and discussion of the new output and support an updated collective understanding of different approaches for modelling the transfer of C-14 in soil-plant systems and in aquatic environments, providing mutual benefit through the sharing of the new experience.

Need for further research
Potential future activities may include model testing against well-defined datasets potentially available from on-going field investigations and site-specific monitoring at a range of sites. This work could provide support for confidence in assessment results through model validation.

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

Modelling Approaches to C-14 in Soil-Plant Systems and in Aquatic Environments

Report of an International Workshop

Version 5
25 January 2014

SSM 2014:30

PREFACE

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

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

This report presents the results of a range of reviews of C-14 behaviour in the environment and describes presentations and discussions held during an international workshop on 12th to 14th February 2013. The workshop was hosted by SKB. Technical input was provided by a wide range of organisations via presentations and discussions, as described in the report. The financial support provided for the project by ANDRA, EDF, NAGRA, NUMO, NWMO, Posiva, NDA (RWMD), SKB and SSM is gratefully acknowledged. An addendum is included which contains additional information supplied by participants after the workshop. The report is compiled from contributions provided by the participants at the workshop, edited by the Technical Support Team (S Mobbs, K Smith, M Thorne and G Smith).

The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved. All material is made available entirely at the user's risk.

Version History

Version 1.0: Draft workshop and review report prepared by the Technical Support Team based on review and participant contributions, 20 March 2013.

Version 2.0: Final workshop and review report prepared by the Technical Support Team based on participant comments on version 1.0, 6 May 2013.

Version 3.0: Final workshop and review report prepared by the Technical Support Team based on participant and sponsor comments on version 2.0, 15 May 2013.

Version 4.0: Final workshop and review report prepared by the Technical Support Team based on participant and sponsor comments on version 3.0 plus addendum describing additional information, 10 January 2014.

Version 5.0: Final workshop and review report prepared by the Technical Support Team, including amendments to several references. 25 January 2014.

Executive Summary

It has long been recognised that C-14 is present in solid radioactive wastes arising from the nuclear power industry, in reactor operating wastes and, more significantly, in graphite and activated metals that will arise from reactor decommissioning. Its relatively long half-life of 5730 years means that there is the potential for releases of C-14 to the biosphere from radioactive waste repositories. These releases may occur as C-14 bearing gases, especially methane, or as aqueous species, and the C-14 will enter the biosphere from below via natural processes or via groundwater pumped from wells.

Whether released from repositories with gaseous or aqueous species, a particular focus of attention is the behaviour of C-14 incorporated into carbon dioxide evolved from the soil by microbial processes. C-14 bearing carbon dioxide mixes with natural C-14 and stable isotopes of C in the soil and the plant canopy atmosphere with degassing and re-deposition allowing inter-compartment transfers with eventual transfer into plant biomass (primarily through photosynthesis) and thence into the food chain.

Models for C-14 dose assessment were reviewed under the BIOPROTA programme in 2005. Subsequently, a more detailed, quantitative C-14 model comparison exercise was completed under BIOPROTA in 2009, with a follow on project completed in 2011. The 2009-2011 model inter-comparison results show that the conceptualisation of the dynamics of the plant canopy atmosphere strongly influences the calculated plant C-14 concentrations. Some models assume less mixing than others and consequently lead to higher calculated plant C-14 concentrations.

Substantial expertise on carbon biogeochemistry already exists in the fields of plant physiology and aquatic ecology and therefore is it beneficial to draw on this to identify the important issues. A poster describing the BIOPROTA C-14 model inter-comparisons, and plans for the forward programme, was therefore presented at the International Radiocarbon Conference in July 2012, and a paper describing the work has been accepted for publication in Radiocarbon Journal. Further studies on C-14 (including modelling, model inter-comparisons, and experimental work) have been on-going by individual waste management organisations and continued interest was shown in further collaborative work, including assessment of doses to humans due to long-term releases of C-14 to surface water bodies.

Noting the above, an international workshop was held from 12 - 14 February 2013, in Stockholm, hosted by SKB, to enable joint review and discussion of the new output and support an updated collective understanding of different approaches for modelling the transfer of C-14 in soil-plant systems and in aquatic environments, providing mutual benefit through the sharing of the new experience.

The workshop was divided into four sessions: a review of recent studies on C-14 in soil-plant systems; a review of the key features of models and data for assessing doses to humans via consumption of aquatic biota following C-14 long-term release to surface water bodies; a review of overall assessments for C-14; and discussion and suggestions for future research activities. Presentations were given by Eden Nuclear and Environment, IRSN, Mike Thorne and Associates (for LLWR and NDA), SKB, Facilia AB, UECWP, GMS Abingdon Ltd (for NIRS), RadEcol Consulting Ltd, SRC, OSU, Nagra, Aleksandria Sciences (for SSM), and CIEMAT.

The following potential future work activities were identified:

a) The recently developed soil-plant models (e.g. those developed by Facilia for SKB, by Mike Thorne and Associates for LLWR, by IRSN and by a team of consultants for SSM) could be run for well-defined cases and the results compared against each other. The level of agreement could be compared with that obtained in the previous BIOPROTA model inter-comparisons.

b) Ideally, these model comparisons could also be related to specific datasets (potentially those from Cap de la Hague and the University of Nottingham), so that the modelling constitutes a validation test.

c) In the context of wetlands, use could be made of the extensive hydrological and C-14 datasets that are potentially available for a site in Canada. A 3D catchment model of the swamp could be constructed, calibrated against the hydrological data, combined with a C-14 loss model, and used to predict the 3D distribution of C-14 concentrations in the swamp and the evolution of that distribution with time.

d) Models for C-14 transport in streams, rivers and lakes should be identified (or developed) and a model evaluation and inter-comparison performed. Ideally, these models would be evaluated by comparing the results obtained from them with monitoring data from major European rivers upstream and downstream of the locations of active discharges from nuclear power plants and other types of installation (e.g. hospitals, research installations, radiochemical manufacturers).

e) A review could be conducted to determine general conceptual model structures appropriate to C-14 transport in different ecological contexts. This could then form a basis for evaluating the temporal and spatial scales over which assessments should be undertaken, taking into account the requirement of being able to undertake assessments for both humans and non-human biota. Interactions with the BIOPROTA SPACE project, which has a similar objective but is not limited to C-14, would be beneficial.

f) Those organisations with special interest in impacts of releases of C-14 may wish to take account of a new UK research programme funded under the RATE initiative on Naturally Occurring Radioactive Material (NORM). This will be exploring biogeochemical processes in the near-surface zone with a view to generating a nationwide characterisation of the radiation environment. Although this programme will emphasise NORM, it will be of relevance to C-14, e.g. through characterisation of the near-surface microbial regime and microbiologically mediated processes.

This report is provided as a substantive record of the workshop presentations and discussion. An addendum to this report contains information about further on-going research work which became available after the workshop.

CONTENTS

1. INTRODUCTION

It has long been recognised that C-14 is present in solid radioactive wastes arising from the nuclear power industry, in reactor operating wastes and, more significantly, in graphite and activated metals that will arise from reactor decommissioning. Its relatively long half-life of 5730 years means that there is the potential for releases of C-14 to the biosphere from radioactive waste repositories. These releases may occur as C-14 bearing gases, especially methane, or as aqueous species, and the C-14 will enter the biosphere from below via natural processes or via groundwater pumped from wells.

Whether released from repositories with gaseous or aqueous species, a particular focus of attention is the behaviour of C-14 incorporated into carbon dioxide evolved from the soil by microbial processes. C-14 bearing carbon dioxide mixes with natural C-14 and stable isotopes of C in the soil and the plant canopy atmosphere with degassing and re-deposition allowing inter-compartment transfers with eventual transfer into plant biomass (primarily through photosynthesis) and thence into the food chain. Modelling of C-14 transfer through the environment has therefore been a key aspect of radiological assessments of radioactive waste disposal facilities for a number of years.

Models for C-14 were reviewed under the BIOPROTA programme in Sheppard and Thorne [2005]. Subsequently, a more detailed, quantitative C-14 model comparison exercise was completed under BIOPROTA in 2009 [Limer et al., 2009], with a follow-on project completed in 2011 [Limer et al., 2012].

The 2009-2011 model inter-comparison results show that the conceptualisation of the dynamics of the plant canopy atmosphere influences the calculated plant C-14 concentrations. Some models assume less mixing than others and consequently lead to higher calculated plant C-14 concentrations. The major source of uncertainty is therefore related to the identification of conditions under which mixing occurs and isotopic equilibria are established: the openness of the canopy and the wind profile in and above the plant canopy are likely to be key drivers. The study provided information with respect to the workings of the models used by various waste management organisations and, thus, identified where key uncertainties lie and gave some confidence for future model developments and applications. Additional work was needed, however, to determine appropriate values of key parameters; much of the justification for the parameterisation of existing models can be traced back to papers from the early 1990s (e.g. [Amiro et al., 1991; Vuorinen et al, 1989]).

Carbon biogeochemistry is a complex topic. However, substantial expertise already exists in the fields of plant physiology and aquatic ecology, from studying carbon turnover, and there is a need to draw on this to identify the important issues. It was therefore considered beneficial to engage with the wider C-14 community to exchange information on important exchange mechanisms and modelling approaches, and to identify the key issues that need to be addressed. A poster describing the BIOPROTA C-14 model inter-comparisons, and plans for the forward programme, was therefore presented at the International Radiocarbon Conference in July 2012, and a paper describing the work has been accepted for publication in Radiocarbon Journal.

Further studies on C-14 (including modelling, model inter-comparisons, and experimental work) have been funded by individual waste management organisations. This work adds new insights based on new assessment work, taking into account a wider range of references. Joint review and discussion of the new output provides mutual benefit through the sharing of the new experience. This new experience includes, among other inputs, reviews of gas transport within the plant canopy, model implementation and inter-comparison studies, and the use of resistance-analogue models in plant canopy studies [Norris et al., 2011; Wilson, 1989; Wilson and Sawford, 1996; Finnigan, 2000; Baldocchi et al, 1983; Shuttleworth and Wallace, 1985; and Shuttleworth and Gurney, 1990], work undertaken on behalf of Low Level Waste Repository Ltd (LLWR) and work currently being undertaken by SKB as summarised at the BIOPROTA meeting in Nancy [BIOPROTA, 2012].

Apart from C-14 in soil-plant systems, renewed interest has been shown in assessment of doses to humans due to long-term releases of C-14 to surface water bodies. It is therefore also considered timely to develop an updated collective understanding of different approaches, for example, going beyond the use of simple concentration ratio approach for incorporation into fish, by using dynamic foodchain models, as discussed some time ago in BIOMOVS II [1996] and more recently in Kumblad et al. [2006].

Noting the above discussion, a project was organised within the BIOPROTA collaboration programme (www.bioprota.org) with the following objectives:

- to prepare and present a paper on the BIOPROTA C-14 work carried out to date, and planned in the forward programme, at the 21st International Radiocarbon Conference in July 2012, with subsequent publication in Radiocarbon Journal;
- to organise and document a workshop to identify the key issues relating to modelling of terrestrial and aquatic pathways for C-14; and
- to develop recommendations for future research activities to address these issues.

This report is the report of the workshop, which was held in February 2013.

1.1 OBJECTIVES AND SCOPE OF THE WORKSHOP

The objectives of the workshop were:

- to review wider literature on C-14 in soil-plant systems;
- to review recent model developments on C-14 in soil-plant systems;
- to review key features of models and data for assessing doses to humans via consumption of freshwater biota following C-14 long-term release to surface water bodies;
- to identify the key issues relating to modelling of terrestrial and aquatic pathways for C-14; and
- to develop recommendations for future research activities to address these issues.

1.2 PARTICIPATION

The workshop, hosted by SKB in Stockholm, was attended by 25 participants from 8 countries, representing a range of operators, regulators, researchers and technical support organisations. Participants are listed in Appendix A.

1.3 REPORT STRUCTURE

Section 2 summarises the presentations on terrestrial systems, Section 3 the presentations for aquatic systems, and Section 4 the presentations on composite systems. Section 5 presents the discussion and recommendations for areas of future work. Appendix A contains the list of participants and Appendix B contains an addendum describing further work that became available at the end of September 2013.

2. CARBON-14 IN THE SOIL-PLANT SYSTEM

As mentioned in the introduction, modelling of C-14 in the terrestrial system focuses on the behaviour of C-14 incorporated into carbon dioxide (CO_2) that is released from the soil by microbial processes. C-14 bearing CO_2 mixes with natural C-14 and stable isotopes of C in the soil and the plant canopy atmosphere with eventual transfer into plant biomass (primarily through photosynthesis) and thence into the food chain. C uptake by the root system is significantly lower than the stomatal atmospheric update and is therefore often considered to be of less importance, even for releases to groundwater from underground repositories. The results of the 2009-2011 model inter-comparison [Limer et al. 2011] showed that the conceptualisation of the dynamics of the plant canopy atmosphere influences the calculated plant C-14 concentrations. A paper describing this work has also been accepted for publication in Radiocarbon Journal, in the Radiocarbon 2012 conference proceedings.

Modelling of C-14 in the soil-plant-atmosphere system is an area of active research and development at many organisations, and with many different disciplines involved. Hence, a review of recent developments in studies of the behaviour of C-14 in terrestrial systems has been undertaken. The key findings of the review are presented below and they provided the basis of an introductory presentation by Shelly Mobbs (Eden Nuclear and Environment). Presentations provided by workshop participants relating to recent developments in C-14 in the soil-plant system are also summarised and the discussion points are summarised in section 2.8.

2.1 REVIEW OF MODELLING OF C-14 IN THE TERRESTRIAL SYSTEM

The papers identified for review covered a wide range of topics, including K_d values in soil, CO_2 release from soils, percentage contribution of soil CO_2 to C in plants, dynamics of CO_2 in the canopy atmosphere, atmospheric releases and transfer to plants, and metabolic models for mammals. The papers came from a wide range of disciplines, including global C cycle (global warming), C-14 dating and sources of errors in dates, C-13 discrimination, canopy dynamics for forests, assessment models, carbon capture and storage, and included several reviews. Key findings are presented for a selection of the publications reviewed.

2.1.1 Trumbore [2009] and Trumbore et al. [2012]. Radiocarbon and soil carbon dynamics

Globally, soils and surface litter store 2-3 times the amount of C present in atmospheric CO_2. Soil respiration integrates the below-ground plant and microbially derived CO_2 and is one of the largest transfers in the global C budget. Understanding of the physical, chemical and biological factors that allow organic matter to persist in and be lost from the soil environment (the age distribution of C in plant material and soils) is needed to understand terrestrial feedbacks to global warming and the changes of soil C over the next century. The 2009 paper summarises the mechanisms for stabilising carbon in the soil and describes the state factor approach for extrapolating from single soil profiles to landscapes. On sloping landscapes, the vertical profiles are influenced by plant production and decomposition rates (which may vary with slope position) and lateral movement of minerals. The C content of soils at the bottom of a slope is higher than that on the eroding slope.

C-14 is one of the few tools that can be used to study the dynamics of the soil-plant–atmosphere on decadal to millennial timescales. The two sources (cosmogenic and weapons testing in the 1950s and 1960s until the atmospheric weapons test ban of 1963) enable it to be used as a dating tool, as a source tracer, and as an indicator of the rate of change of carbon in reservoirs. In particular, comparison of the radiocarbon signature of decomposition-derived CO_2 with the time history of C-14 in the atmosphere provides a measure of the time elapsed between C uptake by photosynthesis and its ultimate return to the atmosphere from terrestrial ecosystems. Changes in the C-14 of C in soil organic matter (SOM) since 1960 show that soil has several intrinsic timescales of accumulation and decomposition. Soil carbon pools that have been stable for centuries to millennia are susceptible to abrupt change when soils cross pedogenic thresholds associated with climate change, changes in vegetation/land use and nutrient input.

Recycling of C-14 through microbial biomass means that organic matter can be old in terms of its C-14 content and hence the C-14 signature of respired CO_2 reflects C substrates of different ages, giving C-14 dates that are too old. This is particularly an issue for radiocarbon dating scientists.

The 2012 paper found that the C-14 signature of respired CO_2 ranges from <1 year (annual grasslands) to >50 years (in tundra). Radiocarbon signatures in CO_2 respired from surface litter layers are similar to the C-14 signatures of the organic matter being decomposed. In contrast, the radiocarbon signature of CO_2 derived from mineral soils is often older than that being respired from the surface litter, and has no relationship to the C-14 signature of bulk soil organic matter.

The radiocarbon signature of respired CO_2 provides a direct test for comparison with carbon cycle models that include plant allocation and decomposition: the 2012 paper describes a comparison with the predictions from the Carnegie-Arnes-Stanford Approach (CASA) model.

C-14 is an excellent, but under-utilised tool to determine the stability of C in SOM. However, the continued decline of C-14 in the atmosphere means that it will become increasingly difficult to use: time is running out for C-14.

2.1.2 Kuzyakov [2011]. Linking C pools with CO_2 fluxes in soil

This paper presents a review of experimental approaches enabling soil C pools to be linked with CO_2 flux from the soil. Understanding pools and fluxes in soil is important because soil stores most of the terrestrial C, and because in most global models soil still remains a “black box” that cannot be used to predict changes under new environmental conditions. Despite the importance of carbon (C) pools and CO_2 fluxes in terrestrial ecosystems, assigning fluxes to specific pools remains unsolved. Interestingly, scientists investigating pools are not closely linked with scientists studying fluxes. The pools reflect the static components of a system, and the fluxes are responsible for its dynamics. Thus, pools and fluxes are responsible for the stability and flexibility, respectively, of any ecosystem.

The background, advantages and shortcomings of uncoupled approaches (measuring only pools or fluxes) and of coupled approaches (measuring both pools and fluxes) are evaluated and their prerequisites – steady state of pools and isotopic steady state – described. The uncoupled approaches include: (i) monitoring the decrease of C pools in long-term fallow bare soil lacking C input over decades, (ii) analysing components of CO_2 efflux dynamics by incubating soil without new C input over months or years, and (iii) analysing turnover rates of C pools based on their ^{13}C and ^{14}C isotopic signature. The uncoupled approaches are applicable for non-steady state C conditions only and have limited explanatory power. Coupled approaches enable the direct linking of pools with fluxes and they work under steady state conditions – with continuous input of new C. Coupled approaches involve the application of tracers and include: (a) continuous labelling e.g. ^{13}C of C pools and CO_2 efflux from soil after C3/C4 vegetation[a] changes or in Free Air Carbon dioxide Enrichment (FACE) experiments, (b) pulse labelling e.g. addition of ^{13}C or ^{14}C labelled organics, and (c) bomb ^{14}C.

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[a] C3 carbon fixation is a metabolic pathway for carbon fixation in photosynthesis. C3 plants are the common plants which open their stomata during the day to breathe in CO_2 and release O_2. They cannot survive in hot climates whereas C4 and CAM plants have adaptations that allow them to survive in hot and dry areas. The isotopic signature of C3 plants shows a higher degree of ^{13}C depletion than the C4 plants.

The underlying assumption is that (1) the amount of C mineralised to CO_2 is proportional to the decomposition rates and the pool size, and (2) various pools in soil contribute in parallel (independently, i.e. without interactions) to the CO_2 efflux with different rates: most studies consider two components. The pools frequently having similar decomposition rates are litter of trees and soil microbial biomass, although the biochemical nature of the pools, their origin, as well as their contribution to various fluxes are completely different.

An example of the abrupt approach is to measure the isotopic signature of SOC and CO_2 flux from soil over a time period following the change (tracer input or vegetation change) and then determine the contribution of two SOM pools to CO_2 fluxes using the relative turnover rates of old and new SOM pools. In the absence of experimental data over a long time period (needed to link pools with fluxes), a simulation was performed. The future challenges include combining two or more approaches to elucidate more than two C sources for CO_2 fluxes, e.g. combined tracers or tracers with C3 to C4 vegetation change.

2.1.3 Bruggerman et al. [2011]. Carbon allocation and carbon isotope fluxes in the plant-soil-atmosphere continuum: a review

This paper provides a review of carbon fluxes based on carbon isotopes studies. The first part of the review considers fractionation processes during and after photosynthesis. Then the review elaborates on plant internal and plant-rhizosphere C allocation patterns at different time scales (diurnal, seasonal, inter-annual), including the speed of C transfer and time lags in the coupling of assimilation and respiration, as well as the magnitude and controls of plant-soil C allocation and respiratory fluxes. The third part of the review considers below-ground C turnover, focusing especially on above- and below-ground litter inputs, soil organic matter formation and turnover, production and loss of dissolved organic C, soil respiration and CO_2 fixation by soil microbes. Plant controls on microbial communities and activity via exudates and litter production as well as microbial community effects on C mineralization are also reviewed. The physical interactions between soil CO_2 and the soil matrix, such as CO_2 diffusion and dissolution processes within the soil profile are also described and, finally, the state-of-the-art of stable isotope methodologies and their latest developments are discussed.

The flux of CO_2 between the atmosphere and the terrestrial biosphere and back is approx. 15–20 times larger than the anthropogenic release of CO_2 [IPCC, 2007]. This large bidirectional biogenic CO_2 flux has a significant imprint on the carbon isotope signature of atmospheric CO_2 [Randerson et al., 2002], which in turn helps to understand the controls of CO_2 fluxes and to predict how they will respond to global change. There is a lack of knowledge on how plant physiological as well as soil biological, physical and chemical processes interact with and affect ecosystem processes, such as net ecosystem primary production and carbon sequestration as well as the larger-scale carbon balance.

Due to the slight difference in atomic mass, physical and chemical properties of substances containing different stable isotopes (such as $^{12}CO_2$ and $^{13}CO_2$) vary, resulting in different reaction kinetics and thermodynamic properties. These result in the “preference” of chemical and physical processes for one over the other (e.g. preference for $^{12}CO_2$ over $^{13}CO_2$) and hence in so-called *fractionation* events, which change the isotopic composition of compounds involved in such processes. The carbon isotope composition is usually expressed in δ notation (in ‰ units), relative to the international standard Vienna Pee Dee Belemnite (VPDB). The carbon isotopic composition $\delta^{13}C$ of any sample is thus expressed as deviation from VPDB:

$$\delta^{13}C=(R_{sample}/R_{VPDB})-1$$

where R_i is the isotope (abundance) ratio ($^{13}C/^{12}C$) and $R_{VPDB} = 0.0111802$.

The notation for isotope fractionation is Δ. Carbon isotope discrimination ($\Delta^{13}C$) is defined as the depletion of ^{13}C during any process preferring the lighter isotope:

$$\Delta^{13}C = (\delta^{13}C_s - \delta^{13}C_p)/(1+\delta^{13}C_p)$$

where $\delta^{13}C_s$ is the carbon isotope signature of the source (e.g. CO_2 when photosynthetic fractionation is considered) and $\delta^{13}C_p$ is the isotopic signature of the product of a process.

An overview of the processes and factors determining the isotope signature of C pools and fluxes is given in

Figure 2-1, taken from Bruggerman et al, [2011].

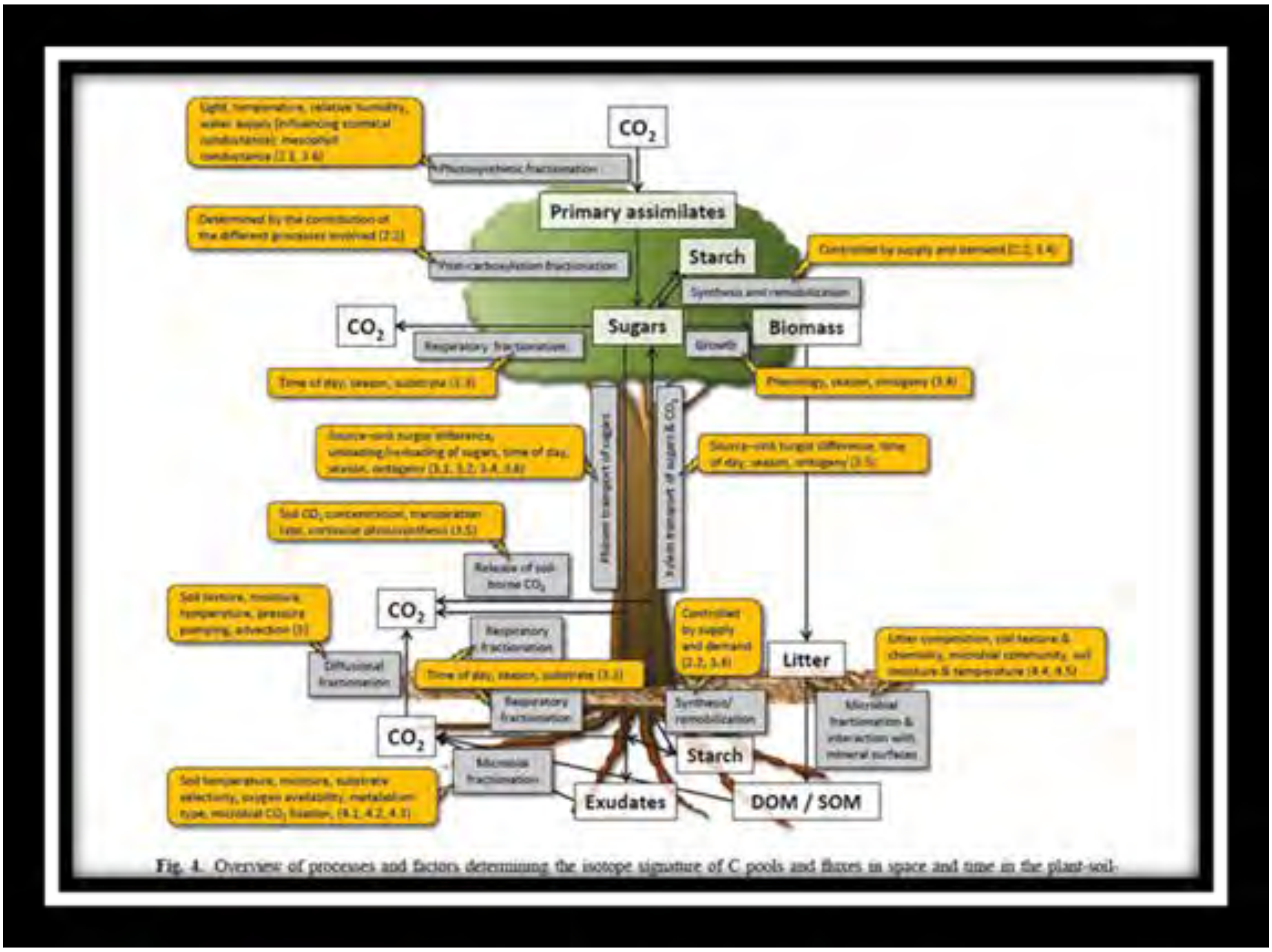

Figure 2-1. Overview of processes and factors determining the isotope signature of C pools and fluxes in space and time in the plant-soil-atmosphere continuum. White boxes represent pools, gray boxes show fractionation or other processes determining the C isotope composition of the involved compounds, and orange boxes depict control factors.

Plant respiration is not fuelled by a homogeneous substrate, but by several C pools with different turnover times and metabolic histories. In rye grass, only 43% of respiration was directly driven by current photosynthates, thus pointing to the importance of short-term storage pools with half-lives of a few hours to more than a day. Below-ground plant parts are supplied by both recent photosynthates and C reserves.

Soil CO_2 efflux is dominated by two major sources of soil respiration: an autotrophic component (roots, mycorrhizal fungi and other root-associated microbes dependent on recent C photosynthates) and a heterotrophic component (organisms decomposing soil organic matter). On average, they contribute equal amounts to total soil respiration, ranging from 10 to 90% in single studies.

A large fraction of C fixed by plant photosynthesis is allocated below-ground: (1) invested into biomass or respired by roots; (2) released as exudates and allocated to soil microorganisms in the rhizosphere[a]; or (3) incorporated as litter into soil organic matter that may be respired by heterotrophic soil microorganisms. Overall, the C flux to soil biota in the rhizosphere is large and C is typically lost from the system within days to months. Environmental conditions imprinted in the $\delta^{13}C$ of photosynthates are thus translated through organisms in the rhizosphere and remain detectable in the autotrophic part of soil respiration. Radiocarbon analysis of root-respired CO_2 showed that roots partly respire older C, indicating that root C stores might serve as respiratory substrates and allow maintenance of respiration rates, at least temporarily.

2.1.4 Werner et al. [2012]. Progress and challenges in using stable isotopes to trace plant carbon and water relations across scales

The paper describes recent progress in understanding plant carbon and water cycling, and their interactions with the atmosphere. It describes how stable isotope studies are a powerful tool for tracing biogeochemical processes across spatial and temporal scales. It provides a useful summary of processes and equations and gives examples covering the leaf scale to the regional scale. Progress and challenges in isotope effects are described for leaf-level processes (CO_2 and H_2O exchange, post-carboxylation and respiratory fractionation, bulk leaf tissue $\delta^{13}C$ and $\delta^{18}O$ and water use efficiency), ^{13}C and ^{18}O isotopes to trace plant integrated processes and plant-soil coupling, community-scale processes, the use of stable isotopes to disentangle ecosystem exchange processes, and regional-scale isotope variation in precipitation and linkages to carbon cycling. New technical and methodological developments in stable isotope research are described and the paper concludes that we are in the midst of a rapid growth in process-based understanding of the behaviour of carbon and oxygen stable isotopes in organisms and in the environment.

2.1.5 Booth et al. [2012]. High sensitivity of future global warming to land carbon cycle processes

Uncertainties in future global warming modelling are usually assumed to arise from uncertainties either in the amount of anthropogenic greenhouse gas emissions or in the sensitivity of the climate to changes in greenhouse gas concentrations. Previous modelling has indicated that the relevant carbon cycle uncertainties are smaller than the uncertainties in physical climate feedbacks and emissions. This paper describes the results of a fully coupled climate–carbon cycle model and a systematic method to explore uncertainties in the land carbon cycle feedback, for a single emissions scenario. The model was run with variations in parameters relating to photosynthesis and soil respiration, and the results indicate that the plausible range of climate–carbon cycle feedbacks is significantly larger than previously estimated. The sensitivity of photosynthetic metabolism to temperature emerges as the most important uncertainty: graphs of the predicted increase in CO_2 showed a strong negative correlation with the optimum temperature of photosynthesis (T_{opt}). Broadleaf trees represent large tropical carbon stocks, where temperatures are most likely to exceed T_{opt} values in a warming climate. Values of T_{opt} for broadleaf trees range between 19^o and 39^o with over 90% within the 28^o-38^o range sampled by this study. To understand future responses of ecosystems to climate change,

[a] The rhizosphere is a narrow zone in the vicinity of the roots characterised by the presence of mycorrhizal fungi and other rhizosphere microorganisms.

consideration of the extent to which plants can acclimatise both photosynthesis and respiration to increasing temperatures is critical. There is an urgent need for better understanding of plant photosynthetic responses to high temperatures, as these responses are shown here to be key contributors to the magnitude of future change.

2.1.6 Le Dizès et al. [2012], Aulagnier et al. [2012], Aulagnier et al. [2013]. TOCATTA and TOCATTA_X models.

The Le Dizès et al. [2012] paper describes TOCATTA, a dynamic compartment model developed at IRSN. The model is implemented in the SYMBIOSE modelling platform and is designed to describe C-14 transfer in agricultural systems exposed to spray irrigation with contaminated water and/or atmospheric C-14 emissions from nuclear facilities operating under normal or accident conditions. The Aulagnier et al. [2012] paper compares TOCATTA with another model PASIM. This led to the development of TOCATTA_khi (TOCATTA_X), based on TOCATTA and including elements of PASIM, as described in Aulagnier et al. [2013]. The models have been tested using the results from field measurements in the vicinity of AREVA NC nuclear reprocessing plant in France.

TOCATTA, TOCATTA_X and the comparison with field data are described in more detail in the presentation by Severine Le Dizès in Section 2.2. Key findings and discussion points are described here.

The TOCATTA conceptual model is given in Figure 2-2. The diagonal elements represent the compartments and the off-diagonal elements are the processes included in the model. The formulae and parameter values used in the model testing are described in the report.

The soil-plant system : conceptual model

SOURCE		Wet Input to Soil				
	CANOPY ATMOSPHERE				Net primary production	
	Volatilisation	SOIL WATER				Migration
	Volatilisation		SOIL AIR			
			Total respiration	SOIL ORGANIC MATTER Decomposition		
				Litterfall Root exudation	PLANT DRY MATERIAL *Biological growth*	Grazing or Cut (for grass only)
						SINK

Figure 2-2 Conceptual model for TOCATTA

The key features of the model are the daily time step and the inclusion of plant growth, using predefined growth curves. A single plant compartment is used, and no root uptake is included in the model. The turnover of soil organic matter (SOM) is based on the widely used Rothamsted C model, using monthly time steps. Volatilisation (transfer of inorganic carbon in soil to the soil-canopy atmosphere interface) is modelled using the simplification proposed by Sheppard et al. [2006]. This requires a value for the plant canopy dilution factor (fraction of C fixed by plants from soil as opposed to that from the above-canopy atmosphere). This factor depends on the area of contaminated soil and the crop height and density; the value of 0.3 used in the model is the value suggested by Sheppard et al. [2006]. Migration due to the movement of soil water to deeper horizons (SINK) is represented by advection only. The input to the model is the daily atmospheric $^{14}CO_2$ concentration, monthly climate data (temperature, rainfall) and the C-14 concentration in irrigation water and monthly irrigation depth, if irrigation is considered.

The model was tested by comparison with an extensive set of field measurements (covering the period from October 2006 to July 2008) made specifically for this purpose in the Validation of TOCATTA (VATO) study. Monthly measurements of soil and grass were made on a rye grass field plot 2km downwind of the AREVA NC plant. Hourly measurements of the Kr-85 activity in air 1.5 m above the plot were used to estimate the daily C-14 atmospheric concentration as $^{14}CO_2$ at the plot since C-14 and Kr-85 are released together from AREVA NC. This is an important dataset and it is hoped that it will be made available to others.

The observed C-14 specific activity in grass shows two peaks above 1000 Bq kg^{-1}: one in November 2006 and one in June 2007. Other lower peaks are also seen (see Figure 2-6) and the mean value from 2006 – 7 is the same as the background for areas away from industrial influence (240 ± 2 Bq kg^{-1}). The grass peaks correspond to, or are subsequent in time to, peaks in the atmospheric C-14 concentration whereas the specific activity in soil is almost constant. Hence the AREVA NC release (rather than the CO_2 from the soil) dominates the plant response.

Comparing the model predictions with measurements (see Figure 2-6) it was found that the specific activity in soil is almost constant and fits well with observations (note that the model was set up with the initial measured C-14 activity in soil, about 420 Bq kg^{-1}). The predicted variations in specific activity concentration in grass were smoother and with lower peaks than measured, although the low values of activity concentration in grass (March 2006, August to October 2007) fitted the experimental data. As a result, refinements were proposed for the model to make it more process based and to use hourly data, in particular to have more than one plant compartment.

Following on from this, the ability of another model, PaSim, to reproduce the observed temporal variability in grass C-14 activity in the vicinity of AREVA NC La Hague was investigated, and the results compared with TOCATTA. PaSim is a pasture model for simulating grassland carbon and nitrogen cycling. It is process based and contains three plant compartments: substrate, shoot and root dry matter. Both TOCATTA and PaSim tend to under-estimate the magnitude of observed peaks in grass C-14 activity, although they reproduce the general trends. In PaSim, shoot growth draws on the C-14 activity in the substrate pool. Plant C-14 activity concentrations can thus be calculated from moving average concentrations in the substrate pool on the basis that older structural dry matter will gradually be replaced by "fresh" structural matter. The averaging period could be regarded as a mean turn-over time for C-14 within the plant. A mean turn-over time for C-14 within the plant was defined, based on both experimental data and the frequency of cuts. It was noted that any averaged signal, with a moving average period ranging from 10 to 30 days, performs better or at least as well as the C-14 activity simulated by either PaSim or TOCATTA and this may be related to the fact that the grass was cut every month. Using a moving averaging period of 15 -20 days for the C-14 activity in the substrate pool in the PaSim model gave the best fit to the measurements.

Therefore a new version of TOCATTA that runs on a sub-daily basis and accounts for sub-daily processes was developed, called TOCATTA_khi (TOCATTA_χ). This version of TOCATTA is based on short-term non-equilibrium and the fast kinetics in the substrate pool (equivalent to sap), as represented by PaSim. Hence, the model calculates the biomass density and growth rate over each time step. It contains less detail than PaSim and is therefore suitable for use in assessments. This model has at least two major advantages: first it reacts completely differently to a release occurring at night or during the day (in agreement with plant physiology); second it is not influenced by the initial conditions for more than the mean turn-over time for C-14 within the plant (e.g. 15 days). The mean turn-over time for C-14 within the plant can be adapted to the management of the grass field (e.g. regarding the frequency of cuts or the grazing regime).

The most recent paper [Aulagnier et al., 2013] describes the new model TOCCATTA_χ in detail and tests it against the measured C-14 activity concentrations in grass in the AREVA NC dataset described above. TOCATTA_χ, using an averaging period of 20 days, performs better than TOCATTA, see Figure 2-6. The atmospheric C-14 activity concentration above the grass was also estimated from hourly ^{85}Kr release rates from the stack by using an atmospheric dispersion model and meteorological data. This was then used to evaluate the uncertainty in the model predictions of activity in grass arising from uncertainty in the estimation of the atmospheric concentrations.

TOCATTA_χ was also tested against a second dataset consisting of monthly grass C-14 activity concentrations recorded over 2005 and 2006 in five unmanaged grass fields located between 1 and 1.6 km from the reprocessing plant. The atmospheric C-14 activity concentration above each grass field was not measured, so it was estimated from the hourly ^{85}Kr release rates, as above. The grass C-14 activity concentrations at some of these fields are more variable than those at other fields and more variable than the first dataset. This is likely to be due to a different management of grass: in the first dataset the pasture is harvested every month, whereas in the second dataset, the grass fields are poorly managed and rarely grazed. The mean turn-over time for C-14 within the plant (represented by the averaging period) used in the model was 40 days for the unmanaged field (consistent with a value of 40 to 100 days for grassland in the literature), and this can be considered to be equivalent to pasture. The uncertainty associated with the grass C-14 activity concentrations simulated by TOCATTA_χ was estimated to be larger than the standard deviation of observations, due to the propagation of uncertainties in the atmospheric activity concentrations above the field.

Although the TOCATTA and TOCATTA_χ models were not developed for waste disposal situations where the activity concentration of the source (in the soil) varies slowly, they provide information that can be used to refine waste management models.

2.1.7 LLWR [2011] and C-14 gas modelling for LLWR in 2012

Recent developments are described in detail in Mike Thorne's presentation, see Section 2.3. Key points are given here.

The 2011 LLWR Environmental Safety Case calculations for C-14 labelled gas are described in LLWR [2011]. This assessment assumed that all C-14 labelled gas released from the near field diffuses through the site engineered cap to the soil zone, and that all C-14 labelled methane that enters the soil zone is converted to C-14 labelled carbon dioxide. A new model for the uptake of C-14 by plants was developed, building upon the RIMERS and enhanced RIMERS models and recent developments in the BIOPROTA forum. The biosphere model contains the soil layer, the canopy layer and the upper layer and is described in Limer et al. [2012]. Within the canopy layer, molecular diffusion occurs and the vertical velocity is zero. In the upper layer, which sits within the upper region of the plant canopy and also the “free” air above, turbulent mixing is included and the vertical velocity varies with height above the zero displacement plane (z_d)[a]. The horizontal velocity was zero below z_d, equal to the friction velocity within the canopy, and varies with height above the canopy. The thickness of these layers, and the degree of plant uptake of carbon from these layers, depends on the canopy density, which will affect the light intensity and thus the rate of photosynthetic uptake of carbon in the canopy profile (P). Using values from the literature, P was given by:

$$P = P_0 \exp(-0.4 \ K \ LAI)$$

[a] The height above the surface where the wind speed is taken to fall to zero. This is assumed to lie within the canopy.

Where

P_0 is the rate of photosynthetic uptake at the top of the canopy

K is the extinction coefficient for diffuse light

LAI is the leaf area index

Five plant types were considered in the model and root uptake was ignored. The study showed that, for a given flux of C-14 labelled gas entering a specified area, the human dose from inhalation is typically five orders of magnitude lower than the dose arising from the ingestion of contaminated crops.

Further refinement of the model occurred in 2012 and is detailed in Section 2.3. The new model is a 1D vertical model with a total of six compartments: plant above-ground, plant below-ground, canopy, above-canopy, soil solution, and soil gas. The structure is given in **Figure 2-7**. The use of a 1D model, a change from the previous model that included horizontal transport within the upper part of the canopy, was justified on the basis that vertical transport would dominate over horizontal transport in the canopy. Average timescales for vertical transport (<16 s) correspond to horizontal movement of <24 m, even for sparse vegetation, and this is insignificant in comparison to the scales expected for releases from disposal facilities.

An important refinement is the inclusion of turbulent mixing within the full depth of the plant canopy, and the variation of the vertical velocity as the plant develops. This refinement is based on a resistance analogue model and increases the exchange between the canopy compartment and the above-canopy compartment. Recycling from plant to soil was excluded since it was argued that plants only incorporate <4.6% of soil respired CO_2 and hence recycled C from root respiration, leaf litter etc. would be insignificant for waste management situations where the source is constant for a long period of time. However, direct root uptake was included in the new model.

Another refinement is the move to a process-based model that includes the growth of the plant. Light-dependent uptake is included and the plant uptake varies over the growing season. However, sensitivity studies showed that the inclusion of plant uptake over the growing season rather than just assuming that it reflects the uptake at maturity has little effect on the results. The most important parameters were found to be the wind speed and the fraction of plant carbon arising from root uptake.

This is a model that is designed for waste management disposal assessments. It would be good to see how it compares with other models and with measurements and whether the inclusion of advection and bioturbation in the soil would influence the results.

2.1.8 Atkinson et al. [2011], Atkinson et al. [2012a,b]. NDA experimental studies.

These experiments are also described in Mike Thorne's presentation, see section 2.3. Key points are given here.

Assessment studies have shown that production of radioactive gases, including $^{14}CO_2$ and $^{14}CH_4$, will occur in geological radioactive waste repositories for intermediate-level wastes and a portion of these gases will migrate to the surface environment over periods of several thousand years. The gas most likely to reach the surface is $^{14}CH_4$. NDA have commissioned some experimental studies which focus on the behaviour and fate of $^{14}CH_4$ introduced into subsurface soil and its subsequent incorporation into vegetation under field conditions. Atkinson et al. [2011] and Atkinson et al. [2012a] describe the first two laboratory experiments. These pilot experiments were designed to explore and develop suitable methods for injection and sampling small quantities of $^{13}CH_4$ in larger-scale experiments. They involved the establishment of small (50 cm high × 15 cm diameter) soil columns in the laboratory. The re-packed soil columns consisted entirely of top soil (0 – 10 cm) while the undisturbed columns represented the natural gradient of top soil to sub-soil which is evident in the field from the soil surface to 50 cm depth.

Following injection of methane gas with a specific $^{13}CH_4$ isotopic composition at a depth of 45 cm, $^{13}CH_4$ diffused toward the soil surface and out into the free atmosphere. This process was complete within 10 hours for re-packed soil and within 48 hours for undisturbed soil. Hence, diffusion was more rapid in the former than the latter, probably due to an increase in total soil porosity during the re-packing process. Measurements of the vertical profiles of CH_4 in both sets of soil columns prior to gas injection indicated that the concentration of CH_4 in the free atmosphere above the columns was higher than the concentrations measured in soil gas samples. This is typical of oxic soils and indicates that microbial oxidation within the soil gives rise to 'consumption' of atmospheric CH_4, hence a net flux of atmospheric CH_4 from atmosphere to soil is observed. The experiments with the repacked soil indicated out-gassing of both CH_4 and CO_2 from the soil columns to the atmosphere when the soil was relatively wet (25-33 % moisture content by volume), demonstrating the highly responsive nature of methanogenesis to soil wetting.

Atkinson et al. [2012b] summarises data obtained from a subsequent field experiment with ryegrass (using the stable isotope ^{13}C as a surrogate for radioactive ^{14}C), and presents numerical simulations of the two laboratory column experiments. The field experiments were carried out from May to August 2011 and provided less consistent results than the column experiments. It was found that the pulse diffuses through the soil column in 8-10 h in vegetated plots, and in 10-24 h in unvegetated plots. This result is consistent with the observations of increased porosity and reduced water content for the vegetated plots compared with the unvegetated plots. The passage times for the CH_4 pulse under natural conditions are also broadly consistent with those observed in the laboratory column experiments, and with those predicted by numerical simulations. A strongly enriched $^{13}CH_4$ signal in the head space 3 hours after injection in the subsoil indicates rapid breakthrough of CH_4 in both vegetated and unvegetated plots. Six hours after injection, the $^{13}CH_4$ signal in the unvegetated plots is higher than in the vegetated plots, consistent with the observation of a slightly slower breakthrough of CH_4 in the unvegetated plots due to reduced effective porosity.

The modelling approach taken was to identify the processes, simulate the various experiments, and then to compare the numerical predictions against the experimental data. Where there were discrepancies between the predictions and the data, the aim was to try to explain those in terms of uncertainties in the numerical model and the data. This approach involves minimal calibration against the experimental data, and is challenging for the numerical models but also informative. There was some uncertainty about the distribution of water in the soil columns and the predicted rates of gas efflux from the soil columns using the model are slower than observed.

2.1.9 Greaver et al. [2005]. An empirical method of measuring CO_2 recycling by isotopic enrichment of respired CO_2

The respiratory based recycling index refers to the flux of respired CO_2 fixed by photosynthesis relative to the total respiratory flux [Sternberg, 1989]. This study measured recycling in a fast growing agricultural cover crop (*C Juncea*) stand using the steady-state model equation which uses a variation of the Keeling plot [Sternberg, 1989]. An empirical method was then used to determine recycling in the same cover crop stand for comparison with the theoretically derived value. This empirical method involved the artificial ^{13}C labelling of respired CO_2 in a treatment plot and comparing the isotopic composition of its respired CO_2 and biomass with those of a control plot. Measurements of $\delta^{13}C$ in root, stem and leaf were taken. The theoretical method gave a value of respiratory based CO_2 recycling of 0.41, while the empirical method gave a value of 0.49. Therefore close to half of the respired CO_2 is refixed during daytime photosynthesis in this densely planted cover crop. Refixation of respired CO_2 during the day should lead to an isotopic enrichment of the remaining respired CO_2

leaving the canopy of the cover crop. Therefore, calculations of gross respiration and photosynthesis using isotopic mass balance equations that do not take this isotopic fractionation into account could be in error.

2.1.10 Sitch et al. [2003]. Evaluation of ecosystem dynamics, plant geography and terrestrial carbon cycling in the LPJ dynamic global vegetation model

The Lund-Potsdam-Jena (LPJ) dynamic global vegetation model combines process-based large-scale representations of terrestrial vegetation dynamics and land-atmosphere carbon and water exchanges in a modular framework. It includes feedback through canopy conductance between photosynthesis and transpiration, and interactive coupling between these fast processes and other ecosystem processes. Ten plant functional types are considered and their fractional cover from year to year is influenced by resource competition and fire response. Photosynthesis, evapotranspiration and soil water dynamics are modelled on a daily time step while vegetation structure and plant population densities are updated annually.

Simulations were run for specific sites where measurements were available and for the world, based on a 0.5^0 x 0.5^0 grid. Seasonal cycles of soil moisture agree well with local measurements and global carbon exchange fields provided a good fit to observed seasonal cycles of CO_2 concentration at all latitudes. The model is being used to study past, present and future terrestrial ecosystem dynamics and interactions between ecosystems and atmosphere.

2.1.11 Lai et al. [1999]. MODELLING VEGETATION-ATMOSPHERE CO_2 EXCHANGE BY A COUPLED EULERIAN-LAGRANGIAN APPROACH

A class of Lagrangian one-dimensional vegetation-atmosphere models (known as CANVEG) successfully combine physiological and biochemical functions derived from leaf-level measurements, radiation attenuation, and canopy microclimate to estimate scalar fluxes (CO_2, H_2O and heat) above the canopy. In such models, velocity statistics, particularly vertical velocity standard deviation (σ_w) and Lagrangian integral time scales (T_L) within the canopy, must be assumed or specified *a priori*. However, these are rarely measured and cannot be specified for an arbitrary leaf area density distribution. Hence a model has been derived to derive velocity statistics within the canopy volume based on leaf area and drag coefficients. The model couples radiation attenuation with mass, energy, and momentum exchange at different canopy levels to estimate mean CO_2 concentration, sources and sinks, and fluxes within the canopy. The paper describes the theoretical basis of the model. A seven-day experiment was conducted in August 1998 to investigate whether the proposed model can reproduce the temporal evolution of scalar (CO_2, H_2O and heat) fluxes, sources and sinks, and concentration profiles within and above a uniform 15-year old pine forest. Measurements of CO_2 and water vapour flux above the canopy were made using an eddy covariance system; measurements of mean air temperature, relative humidity and net radiation were also made at the top of the canopy. A multi-port system was installed to measure the CO_2/H_2O concentrations inside the canopy at 10 levels, see Figure 2-3 .

The model reproduced well the measured depth-averaged canopy surface temperature, and CO_2 concentration profiles within the canopy volume, CO_2 storage flux, net radiation above the canopy, and heat and mass fluxes above the canopy, as well as the velocity statistics near the canopy-atmosphere interface. Implications for scaling measured leaf-level biophysical functions to ecosystem scale are also discussed.

The calculations showed that the magnitude of the CO_2 storage flux relative to the flux measured above the canopy is significant only during the early morning (when the evening CO_2 build up is flushed into the atmosphere) and the late afternoon (when ground efflux and canopy respiration are large yet the atmospheric transport capacity is small). However, the overall contribution of storage flux, when depth and time averaged over the entire experiment duration, was two orders of magnitude smaller than the fluxes at the canopy top. Additionally, the overall comparison between modelled and measured mean CO_2 concentration and canopy fluxes did not significantly improve when the storage flux was included. The mean soil moisture content in the root zone (0–30 cm) was 0.16.

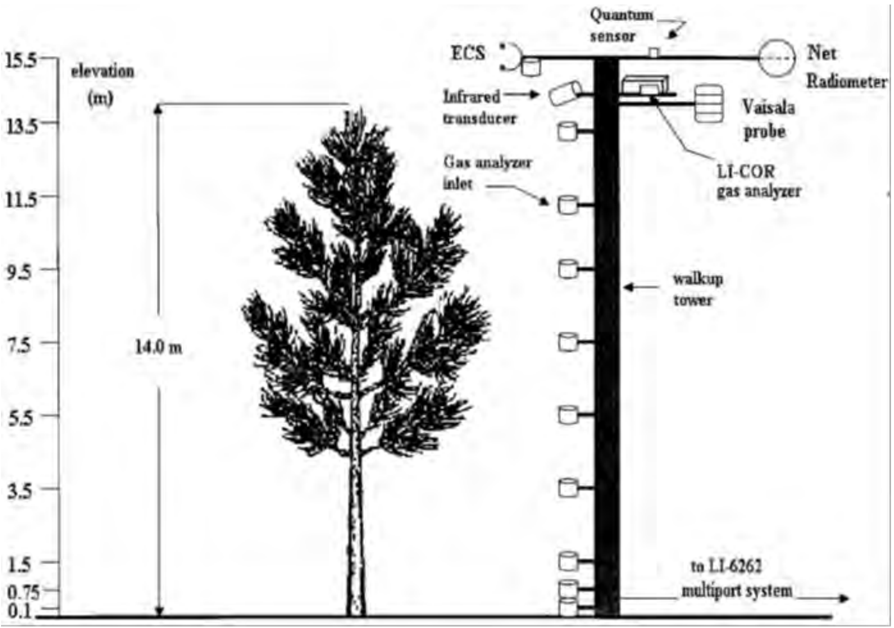

Figure 2-3. Schematic display of the experimental setup for Lai et al. [1999]

2.1.12 Sternberg [1989]. A MODEL TO ESTIMATE CARBON DIOXIDE RECYCLING IN FORESTS USING $^{13}C/^{12}C$ RATIOS AND CONCENTRATIONS OF AMBIENT CARBON DIOXIDE

Carbon dioxide from respiration of forest litter can be dissipated in two ways; photosynthesis and turbulent mixing with the atmosphere. Because there is isotopic discrimination associated with photosynthesis and none with turbulent mixing, different relationships between carbon isotope ratios and concentrations of ambient forest carbon dioxide will occur, depending on which process is responsible for the dissipation of carbon dioxide. A steady-state model predicting the relationship between ambient forest CO_2 concentrations and $\delta^{13}C$ values as a function of the proportion of respired CO_2 reabsorbed by photosynthesis is presented here. Comparisons of the predictions of this model with data collected in a tropical moist forest in Panama show that about 7-8% of the respired carbon dioxide is recycled via photosynthesis.

2.1.13 Root uptake factors given in the literature

The fraction of C in a plant that is obtained from root uptake is calculated from $\delta^{13}C$ values, based on the discrimination between C-12 and C-13 during photosynthesis:

$$\text{Fraction from root uptake} = \frac{(\delta^{13}\text{C in crop-discrimination factor-} \ \delta^{13}\text{C in air})}{(\delta^{13}\text{C in soil-} \ \delta^{13}\text{C in air})}$$

The transfer factor is then given by:

Transfer factor = C in crop/C in soil * fraction from root uptake

A selection of discrimination factors obtained from the literature is given in Table 2.1 and some measured $\delta^{13}C$ values reported in the literature are given in Table 2.2. They illustrate the variation in the reported values of the discrimination factor and $\delta^{13}C$. This should be taken into account when using them to derive the root uptake fraction. Values of the root uptake fraction given in the literature are given in Table 2.3.

Table 2.1. Discrimination factors from the literature

Reference	Discrimination factor
Tagami et al. 2009, Tagami and Uchida 2010	rice -18 to -20‰ and central value is -19‰
Gillon et al. 1997	wheat -15 to -32‰, Bean -16 to -27‰
Farquahar 1989	C3 plants -16 to -23‰
Sternberg 1989	tropical forest -22.8‰
Casper 2005	Cryptantha flava -19.5 to – 20.4‰

Table 2.2. δ13C values from the literature

Reference	$\delta^{13}C$ value
O'Leary 1988	-21‰ to -35‰, wheat -22‰ to-19‰, C3 plant mean -27.1‰
Ford 2007	pine seedling -31.7‰
Trumbore 2009, Phillipson 2012	-25‰
Tagami et al. 2009, Tagami and Uchida 2010	rice -28.09‰ to -26.3‰, mean -27.1‰; wheat -28.5‰; veg -27.7‰
Capano et al. 2012	tree -25‰ to -21‰ except near volcano
Cunningham 2009	-28‰ (5m away from source)

Table 2.3. Root uptake fraction from the literature

Reference	Value for root uptake fraction
Tagami et al. 2009	0 – 1.6%; Transfer factor 0.05 to 0.5 for rice (mean = 0.3)
Yim and Carron 2006	<2%
Ishii et al. 2010	<5%
Ford et al. 2010	1.6% below ground pine seedling
Tagami and Uchida. 2010	rice 0.6%, other 5.5% (up to 7.3%)

2.1.14 Soil respiration and soil reservoir information from the literature

Moisture and temperature affect soil respiration because they affect organic matter decomposition. Moyano et al. [2012] developed a process model based on porosity and bulk density. They plotted respiration against four soil moisture measures for 90 soil types and found a predictable relationship, and that organic and mineral soils differ.

Information on the soil reservoir from various publications is summarised in Table 2.4.

Table 2.4. Soil reservoir information from the literature

Reference	Soil reservoir information
Carbone+Trumbore 2007	3 carbon pools with mean residence times of: 0.5 d (fast); 19.9 d (intermediate); and 9 months (slow). (Above ground C pools: 1 d (fast) and 18.9 d (intermediate)). Respired C mean age: Shrub (3.8 to 4.5d) which is less than that for grass (4.8 to 8.2 d)
Trumbore 2009	SOM fast <1y, weak 10y and slow 1000y, should consider more than 20cm depth in soil
Marzaioli 2012	Mean residence time in top 10 cm 100-4000y
Tagami et al. 2011	Less than 3% remains in soil after 24h
Risk et al. 2002	Concentration of carbon dioxide at the soil surface is 5-8x concentration in atmosphere; respiration is temperature dependent

2.1.15 Other publications

Key points from other publications reviewed are:

- Rice et al. [2010]: methane flux from trees growing in flooded soils is significant;
- Soter [2011]: Recycling of respired CO_2; canopy retards respiration from soil;
- Striegl and Wickland [2001]: CO_2 uptake changes from 29% to 9% of soil respiration as pine forest ages.

2.1.16 Conclusion of review

In conclusion, global warming, global carbon cycle, carbon capture and carbon dating research can produce understanding of the processes involved in the terrestrial C cycle. Detailed models based on these processes have been developed and have been shown to be able to model short-term fluctuations. These detailed models are used to explore the sensitivity of the system to assumptions and hence to inform the development of assessment models which can address the longer timescales of importance to waste disposal.

Additional references cited in the review were: Baker et al. [2000], Conrad R [1996], Baldocchi et al. [1983], Ogiyama et al. [2008], Suarez and Sinunek [1993].

2.2 TRANSFER OF C-14 TO A GRASSLAND ECOSYSTEM

Severine Le Dizès-Maurel (IRSN) presented.

The objectives of the IRSN VATO programme are to:

- Estimate the fluxes of C-14 (and H-3) in a grassland ecosystem (air, rain, grass, soil water) in relation to:
    - Evolution of the concentration in air (day/night);
    - Weather conditions;
    - Land use (grazing, maize silage and hay), and;
- Estimate the transfer of C-14 (and H-3) to cow's milk based on an animal diet; in order to advance the understanding of underlying processes and obtain well-documented data to validate models.

The C-14 experimental programme ran from 2006 to 2009, and the modelling programme from 2009 to 2012. The H-3 programme is now starting and will run from 2013 to 2017, again with an experimental phase followed by (or concomitant with) a modelling phase.

IRSN developed the TOCATTA model for atmospheric releases and/or liquid releases in spray irrigation of C-14. The model represents agricultural systems and can model acute and chronic releases in the form of CO_2. TOCATTA is a dynamic model based on plant biomass growth where the growth curves are either predefined or derived from experimental data. Isotopic equilibrium between the quantity of newly created plant biomass and the surrounding air is assumed at each time step (i.e. 1 day). The model is parameterized for various types of agricultural plants, broken down into three groups: annual crops, vegetable crops and pasture grass. Two categories of soils are considered: sandy soil and clayey soil. The model is integrated in the SYMBIOSE platform so that it can provide an operational tool for environmental survey and assessment around French NPPs.

The experimental programme took place at Atelier Nord, a site downwind of the Areva NC reprocessing plant. The same plot of grass was cut to about 1cm every month simulating grazed grasslands with a recovery period of 1 month. The C-14 atmospheric activity concentrations were obtained from continuous measurements of Kr-85 activity concentrations at the site because of concomitant releases of Kr-85 and C-14.

Figure 2-4 shows results indicating rapid fluctuations of the signal in air and grass due to the wind direction and the operation of the facility and no fluctuation in soil due to a poorly reactive pool of organic matter. The model predicted values are lower (up to 40%), than measured concentrations and the variability between months is underestimated. This underestimation is due to the assumption of daily isotopic equilibrium between the plant and the air, i.e. there is no difference whether a release occurs during the day or during the night. A new model TOCATTA_χ (TOCATTA_khi) has been developed for grass to simulate intra-day C-14 transfer in the soil-plant-atmosphere system. This incorporates the key physiological processes of the PASIM model (photosynthesis and growth, for example), at an hourly time-step, according to local agro-meteorological data. It therefore takes into account the intra-day variability of C-14 releases.

The plant turnover time was derived by fitting the model to the experimental observations: T = 20 days gave the optimum correlation and minimum Root Mean Square Error (RMSE). This implies that the plant would renew its stock in about 20 days, consistent with the frequency of cuts, which is about 30 days. A comparison between the model and observations is given in Figure 2-6.

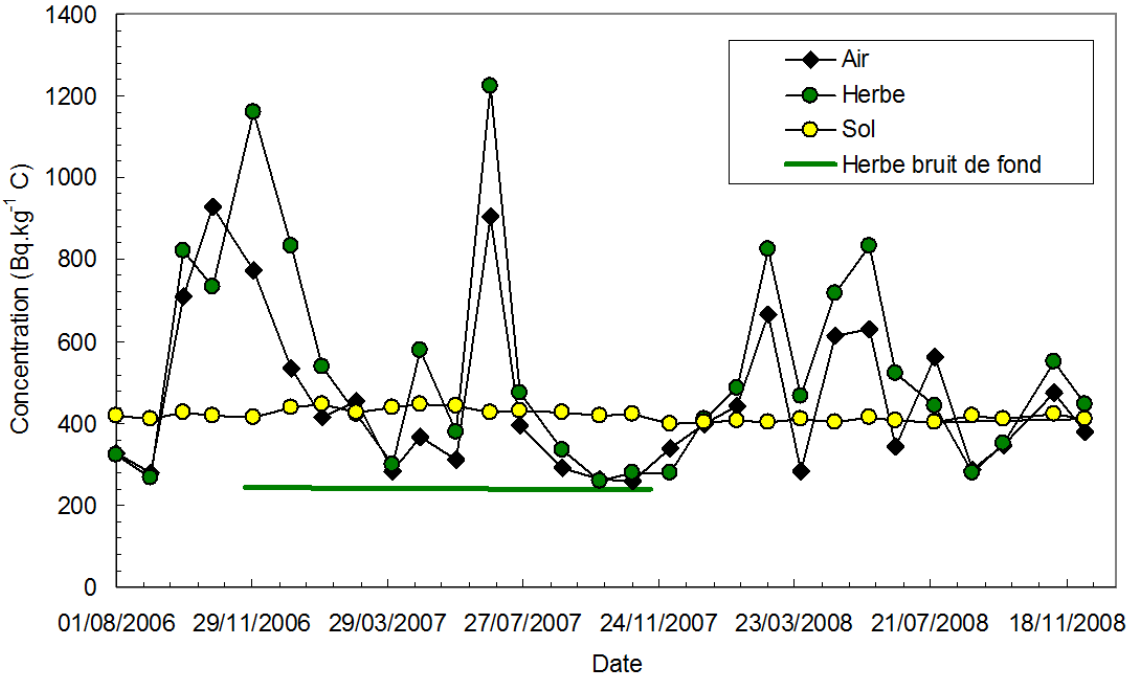

Figure 2-4. C-14 concentration measurements in air, grass and soil

The interaction matrix for the TOCATTA_χ model is given in Figure 2-5.

Plant						
	Organic Matter					
		Shoot (structural dry matter)			Ageing	Cut or grazing
			Root (structural dry matter)		Ageing	
		Biological Growth	Biological Growth	Sap (substrat)		Respiration
					RestOf Plant	
				Photo-synthesis		RestOfWorld

Figure 2-5. Interaction matrix for TOCATTA_χ

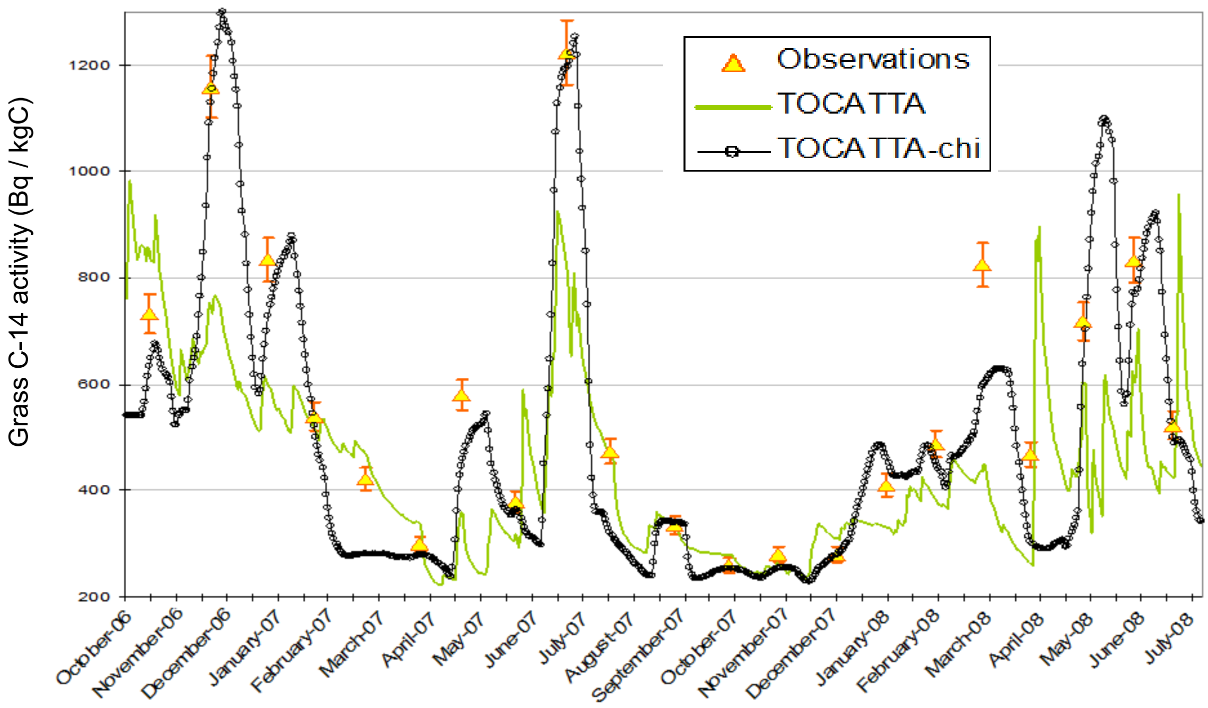

Figure 2-6. Comparison of model and data for Grass C-14 activity (Bq/kg C)

Figure 2-6 shows that TOCATTA_χ is better correlated with observations and provides a better fit to the variability of observations than the TOCATTA model. Hence it is important to adapt the model to time-varying releases and meteorology by using an hourly time-step. Adjustment of the mean turnover time in the plant corresponding to different management modes of the grass was required.

TOCATTA_χ will be used in the modelling phase for the H-3 part of the VATO programme.

The presenter gave the following list of references: [Le Dizès et al., 2012, Riédo et al., 1998, Vuichard et al, 2007, Aulagnier et al., 2012].

2.3 UPDATE ON C-14 MODELLING IN THE UK

Mike Thorne presented on behalf of LLWR and RWMD.

The presentation described a detailed model that LLWR has developed. A simplified assessment model is also used and the simplifications are justified through sensitivity studies undertaken with the detailed model. The structure of the model is shown in Figure 2-7, adapted from Thorne and Walke [2013]; work carried out for LLW Repository Ltd.

The model is a 1D vertical model. The exclusion of horizontal transport of C-14 labelled gas within the plant canopy represents a change from the 2011 model, which included potential horizontal transport of C-14 labelled gas only within the upper part of the plant canopy. The use of a 1D model is justified by considering 10 m as the smallest spatial scale over which lateral migration would be significant. Average timescales for vertical transport through the plant canopy are 0.64, 4.1 and 16 seconds for short grass, open grassland and root crops, respectively. On these timescales, the horizontal displacement, based on wind speeds at the top of the plant canopy, are 0.08, 1.35 and 3.16 m for short grass, open grassland and root crops, respectively. Even if the vegetation is sparse (tending to increase the horizontal wind velocity at the top of the plant canopy), the horizontal displacements will be no more than 0.59, 5.34 and 24.0 m for short grass, open grassland and root crops, respectively.

Horizontal velocities and therefore horizontal displacements within the plant canopies will be substantially lower.

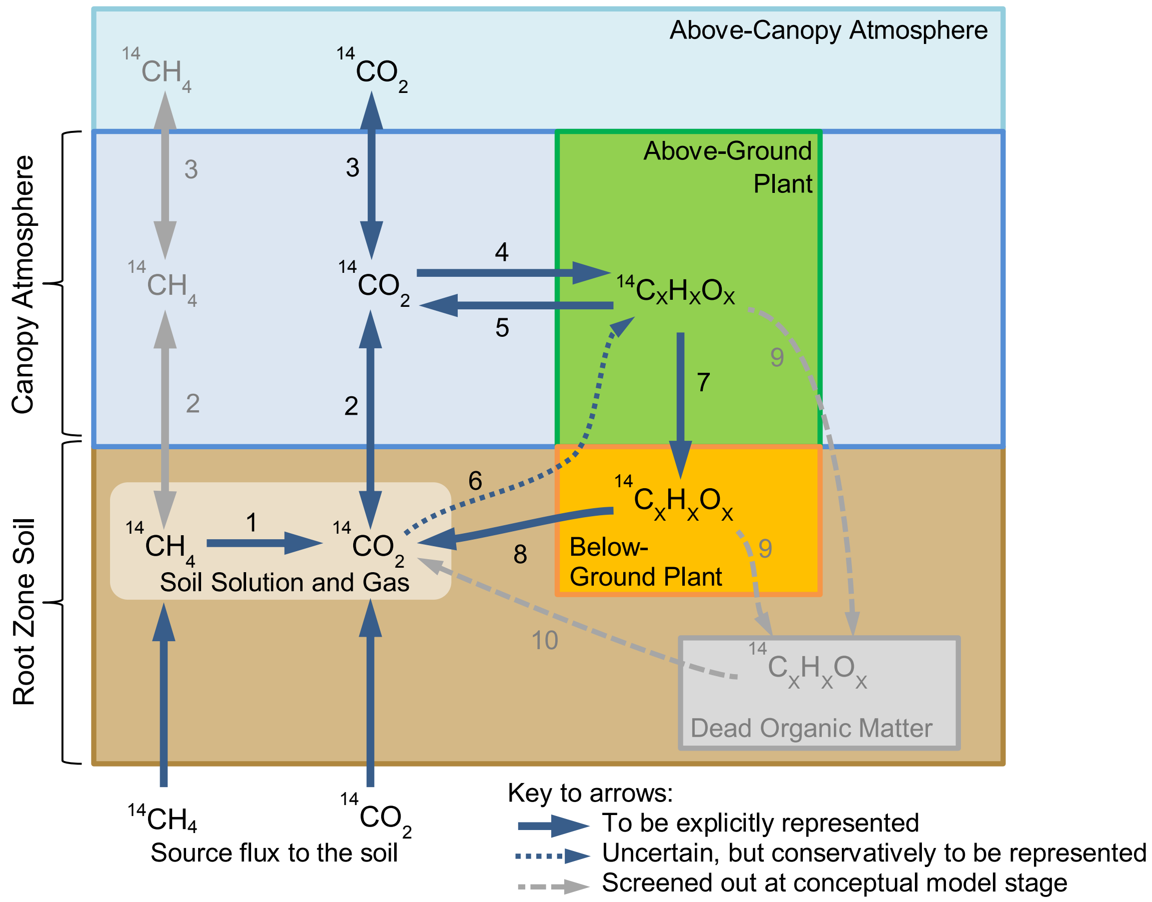

Figure 2-7. Detailed LLWR model

C-14 bearing methane and carbon dioxide are referred to as 14CH4 and 14CO2, respectively. C-14 bearing carbohydrates are referred to as 14CXHXOX. Processes are numbered below:

1. Microbially mediated oxidation of methane to carbon dioxide.
2. Diffusion and atmospheric pumping.
3. Turbulence, diffusion and atmospheric pumping.
4. Uptake via stomata and incorporation into carbohydrates via photosynthesis.
5. Above-ground respiration.
6. Direct root uptake and translocation to the sites of photosynthesis.
7. Translocation to the roots.
8. Root respiration.
9. Direct and indirect incorporation of dead plant material into the soil organic matter.
10. Decomposition.

Turbulent mixing throughout the canopy is included, see Figure 2-8, from Thorne and Walke (2013); work carried out for LLW Repository Ltd. This is a significant development since the 2011 model.

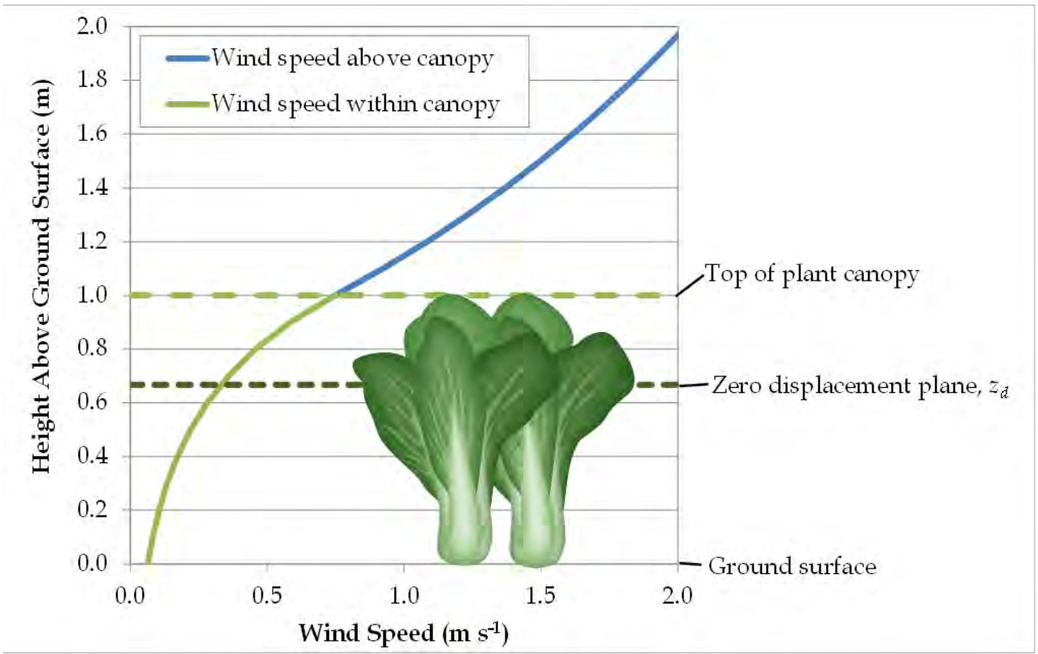

Figure 2-8. Turbulent mixing in the LLWR model

The zero displacement plane is not a boundary where the velocity is zero, but the height at which the velocity gradient changes.

The model includes the variation of uptake into the plant through the growing season as plant morphology and canopy height develop. Initially, there will be little to no canopy coverage of the soil and most of the C-14 released from the soil will be lost directly to the atmosphere. C-14 released to the canopy of seedlings will be subject to higher wind speeds and a greater degree of turbulence than C-14 released to mature canopies, which will provide close to 100% coverage of the soil. The C-14 taken up by plants and incorporated into plant tissues will reflect the integrated uptake over the plant's growing period.

The 2011 assessment conservatively adopted the assumption that the concentration of C-14 within harvested tissues reflects the concentration of C-14 labelled carbon dioxide within the plant canopy at maturity. However, sensitivity studies have demonstrated that the refinement to the model to include uptake through the growing season is of limited significance.

C-14 incorporated into plant material can be returned to the soil by several processes including root respiration, root loss, exudation of organic substances to enhance nutrient uptake and return of dead plant material to the soil. Degradation of soil organic matter by microbes can release associated C-14 to the soil solution. Plants only incorporate about 4.6% (as an upper bound estimate) of the carbon that passes through the plant canopy from the soil into plant tissues. This implies that recycling of C-14 back to the soil need not be explicitly represented in the detailed model, as it would represent a minor fraction of the C-14 flux passing through the soil while the source persists. Hence, recycling from plants to soil is not included in the model.

There is potential for some C-14 in soil solution to be taken up via the plant roots either actively (facilitated by natural mycorrhizae associated with the roots) or passively (with the transpiration flow of soil water). The transpiration flow within the plants has potential to transport this C-14 to the site of photosynthesis in the leaves. There is therefore a conceptual route for potential direct uptake of C-14 from the soil and its incorporation into the products of photosynthesis. The fraction of plant carbon arising from direct uptake of carbon dioxide from the soil will be small, given that plant physiology is designed to take-up carbon from the atmosphere.

There is very little literature available that enables this route to be clearly identified and parameterised, given the difficulty of distinguishing between soil- and atmosphere-derived carbon within plants.

The concept of the photosynthetic discrimination factor (F_{C13}) is important when deriving estimates of root uptake from experimental measurements of $\delta^{13}C$ in plants and soil.

$$x = (\delta C_{crop} - F_{C13} - \delta C_{air})/(\delta C_{soil} - \delta C_{air})$$

hence $F_{C13} = \delta C_{crop} – \delta C_{air} – x(\delta C_{soil} - \delta C_{air})$

where δC_{crop} is the $\delta^{13}C$ value for the crop;

δC_{air} is the $\delta^{13}C$ value for the ambient air (taken as -8.0 ‰);

δC_{soil} is the $\delta^{13}C$ value for the soil;

F_{C13} is the photosynthetic discrimination factor for rice (taken as -18.0 to -20.0 ‰, with a central value of -19 ‰).

The $\delta^{13}C$ in plants is intermediate between that in air and that in soil, with a correction due to the discrimination factor. Looking at a dataset of $\delta^{13}C$ values in rice (ranging from -28 to -26) and $\delta^{13}C$ in soil (ranging from -22 to -28), a root uptake factor of <5% was found to be the best fit to the data: a root uptake factor of >5% resulted in 40% of the values corresponding to a discrimination factor outside the expected range. It is generally accepted that the contribution to plant carbon due to uptake from the roots is up to 1-2%. The reduction in C-14 concentrations in the plant canopy due to turbulent mixing with the wider atmosphere means that the small fraction of plant carbon that may arise from direct uptake from the soil becomes potentially important and hence it is included in the model.

The model for vertical transport of C-14 labelled and non-radio-labelled carbon dioxide from the soil atmosphere to the above-canopy atmosphere adopts an aerodynamic resistance analogy. This type of approach has been used to construct a wide variety of models for simulating processes within the plant canopy and enables the LLWR model to draw on a broad base of literature. The model of Shuttleworth and Wallace is used as the basis for the resistance analogue model adopted for the LLWR assessment. The Shuttleworth and Wallace model is an established resistance analogue model that has been widely supported by comparison against observational data. Resistance analogue models typically start at the ground surface. In the case of the LLWR model, concentrations in the soil are needed, so the resistance model is extended to include the root zone. The associated resistance is estimated based on observed gradients of carbon dioxide concentrations between soils and the canopy atmosphere. In the 2011 ESC, the cautious assumption was made that the exchange between soil and below-canopy atmosphere was determined solely by diffusion. The use of observed gradients in carbon dioxide concentrations between the soil and the open atmosphere effectively means that other processes are implicitly represented, including atmospheric pumping.

Vertical attenuation of light in the canopy is also included in the model. For plants that develop a relatively closed canopy, the fraction of uptake of carbon dioxide from the upper and lower canopy atmosphere is determined by the relative light intensity. This represents a development over the reference model adopted for the 2011 assessment, which distributed uptake uniformly with height.

The model is implemented in a spreadsheet. Output includes plant growth curves over time, aerodynamic resistance and wind velocity curves with time, and the variation of specific activity in deep soil, surface soil, lower canopy atmosphere, upper canopy atmosphere, above canopy atmosphere, above ground plant and below ground plant with time. The sensitivity studies showed that the degree of carbon uptake from the root zone was the most important parameter.

Thus, a comprehensive new model for the behaviour of C-14 in soils and plants has been developed. This builds on work previously undertaken for Nirex and for RWMD in the BIOPROTA programme. It also incorporates important new understanding obtained from literature reviews. The model is much more realistic and substantially less cautious than the Enhanced RIMERS model previously used by RWMD (the results are approximately a factor of 10 lower). The distinction between detailed and assessment-level models has been useful in facilitating detailed quantitative process analyses and in justifying appropriate simplifications that can be made when embedding the model within a larger assessment framework.

Experimental studies designed to determine how much methane is converted to CO_2 are being undertaken by Amec and the University of Nottingham for the NDA. These experiments use methane enriched with ^{13}C i.e. with a distinct $\delta^{13}C$ signature. Phase 1 considered columns in the laboratory and Phase 2 is using soil columns in a field. The enriched methane is injected directly into the soil. The Phase 1 studies demonstrated rapid movement of methane through the soil, taking 300 minutes to come out from 0.4 m depth. The results of the Phase 2 studies will be available in October 2013. These will be much more detailed than the results of the Phase 1 studies, because a new design of soil gas sampler has been used that has a sensitivity about two orders of magnitude greater than that used previously. This has allowed field studies in which the lateral transport of injected gas in the soil has not had to be constrained to be sub-vertical.

2.4 CARBON FORMS AND TURNOVER RATES IN NORTHERN PEAT LANDS / ORGANIC SOILS

Peter Saetre (SKB) presented.

Discharges of deep groundwater in the area around Forsmark are likely to occur in wetlands or lake-mire complexes. Radiocarbon leaking from a geological repository is likely to be in the form of CO_2, CH_4 or LMWOC (low molecular weight organic carbon), but it is primarily in the form of CO_2 that radiocarbon can be fixed by primary producers and thus enter the terrestrial and aquatic food chains. Hence the degradation of organic carbon and the fate of methane in wetlands is an important area of interest. The organic cycle, see Figure 2-9, leads to decreasing decomposability.

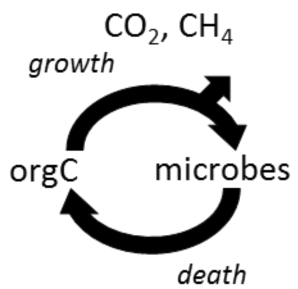

Figure 2-9. Schematic diagram of the organic cycle

The fate of radiocarbon entering peatlands depends on whether there are oxic or anoxic conditions. In anoxic conditions LMWOC can transform into CO_2 by anaerobic respiration or into methane, and in the presence of acetates; in the presence of hydrogen, the CO_2 can be transformed into methane (autotrophic reaction). In oxic conditions LMWOC and CH_4 will be oxidised to CO_2. The conceptual model of the wetland is clay overlain by peat (Catotelm), overlain by peat above the water level (Acrotelm), see Figure 2-10.

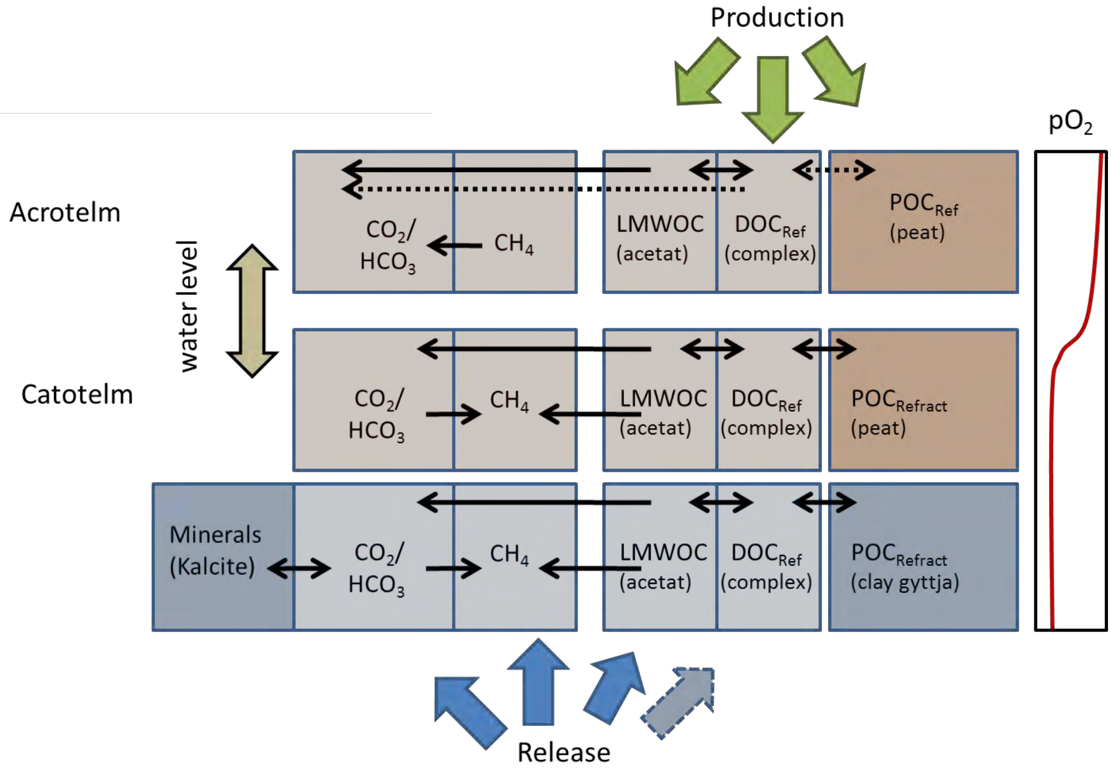

Figure 2-10. Conceptual model for a wetland ecosystem

Concentrations of CO_2, CH_4 DOC and acetate (mg C L^{-1}) in bogs, fens and other wetlands from a literature review were presented. Potential anaerobic C mineralisation rates in Boreal peatlands were estimated from studies of mire types in Michigan (bog, intermediate fen, rich fen) and Finland (ombrotrophic bog, oligotrophic fen, mesotrophic fen) and the turnover time was found to be about 6 days for aceticlastic methane production in the active layer, and about 11 days for autotrophic methane production (from CO_2). Approximately 66.6% of methane flux in nature is from acetate reaction, and >67% in methane-rich environments. However, in oligotrophic environments and for some periods of the year autotrophic methane production may dominate aceticlastic production.

The potential methane production was obtained from a review of results from unrestricted sampling and from active layer sampling studies. The results for Sweden were compared with actual emissions, see Table 2.5. The results indicate that most methane produced in northern peatlands will typically be consumed before it can be emitted to the atmosphere. The net emissions of methane tend to decrease with lower water level as the potential for methane consumption is high in oxic environments.

Table 2.5 Potential methane production compared with emissions

Mire Type	water table (m)[c]	CH_4 Fluxes Production mg m^{-2} day^{-1}	Emission mg m^{-2} day^{-1}	Emiss./Prod.
Northern Sweden[a]				
poor fen, flark	-0.02	340	72	21%
poor fen, hummock	-0.19	300	1	0.3%
short-sedge fen, dry	-0.14	400	-3	NA
short-sedge fen, wet	-0.07	1300	5	0.4%
mesotrophic tall-sedge fen, flark	0.08	1450	70	5%
mesotrophic tall-sedge fen, slope	-0.15	385	8	2%
South and Central Sweden[b]				
sphagnum dwarf shrub (n=41)	-0.32		13 ± 2[d]	
transitional fens (n=4)	-0.37		8 ± 2	
short sedge fens (n=80)	-0.31		29 ± 9	
tall sedge fens (n=43)	-0.23		46 ± 10	

Methane oxidation rates in CH_4 producing and consuming ecosystems were reviewed. In CH_4 producing ecosystems the median area-specific methane oxidation rate (MO) ranged from 3.1 to 4 10^4 mg CH_4 m^{-2} d^{-1}; in CH_4 consuming ecosystems the median MO ranged from 0.55 to 0.97 mg CH_4 m^{-2} d^{-1}. The net flux of greenhouse gases from mire and drained organic soils in Sweden indicates that the methane emission from bogs and fens is limited, but a significant amount from reeds (in the form of bubbles and via aerenchyma[a]).

The degradation half-lives for low molecular weight organic compounds in soil and water under aerobic conditions were reviewed and ranged from 1 to 7 days. The corresponding values for anaerobic conditions were 1 to 28 days. Thus low molecular weight organic carbon (LMWOC) is likely to be oxidized to CO_2 (mineralised) within days in aerobic environments. In anoxic environments, LMWOC is transformed to CH_4 within days to weeks in the active layer, while transformation of CO_2 to methane is somewhat slower.

Potential methane production is one or two orders of magnitude higher than observed emission rates. There is a high potential for methane oxidation in methane-rich environments and this will increase when soil/peat is exposed to elevated CH_4 concentrations. Nevertheless, methane may escape during episodic events by ebullition and through vegetation (aerenchymal transport). Thus whereas biochemical transformation rates will determine the fate of methane dissolved in groundwater, transport (dispersion) and phase transitions (dissolved/gas) must also be considered when assessing the fate of geological methane reaching the biosphere.

2.5 MODELLING ATMOSPHERIC TRANSFER

Rodolfo Avila (Facilia) presented.

[a] spaces or air channels in the leaves, stems and roots of some plants, which allow exchange of gases between the shoot and the root; common in wetland plants.

The work was done for SKB. The assessment context is to model the continuous, long-term release of C-14 as CO_2 from soil over an area covered by grass, crops like cereals (barley) and vegetables. An additional input of C-14 as CO_2 is considered from irrigation water. The assessment endpoints are concentrations in plants and in atmospheric air above the release area.

The mean wind profile above short vegetation (a) and above and within the roughness sub-layer of a forest canopy (b) are shown in Figure 2-11 taken from [Tagesson et al. 2012].

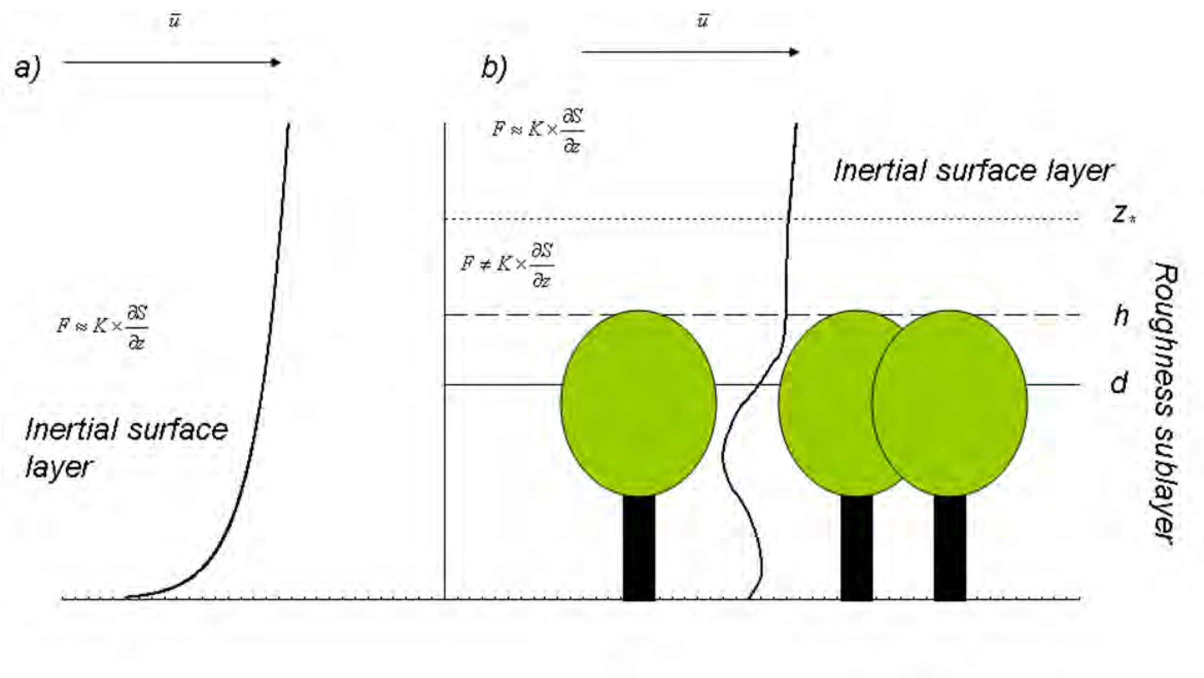

Figure 2-1 (a) Mean wind profile above short vegetation and (b) above and within the roughness sub-layer of a forest canopy, from Tagesson et al [2012]

The wind profile depends on the atmospheric stability and is somewhat complex. The simplest model, assuming that the specific activity is the activity released per unit area divided by the primary production, ignores dilution and is too conservative. A modification to take account of losses due to vertical turbulence can be applied:

$$SA = RRI * \frac{\frac{Release\ rate}{Area}}{NPP}$$

where RRI is the respiratory recycling index (the fraction of CO_2 released from the soil that is assimilated by the vegetation).

In the next development vertical turbulent fluxes were neglected. Instead, a mixing height (hmix) above the canopy was used as the input parameter to estimate dilution with CO_2 from above-canopy atmosphere. A problem with this model lies in the difficulty in estimating this parameter. The wind speed at the vegetation height is used to calculate advective fluxes. It is calculated from the wind speed at a reference height using a simple power dependency of the wind speed with height. Since the uptake of C can take place at lower heights where the wind speed is lower, this model can lead to underestimations of the concentrations, especially for small areas. Assuming a large value of hmix can also lead to underestimations.

The next development of the model was based on micrometeorology and is shown in Figure 2-12.

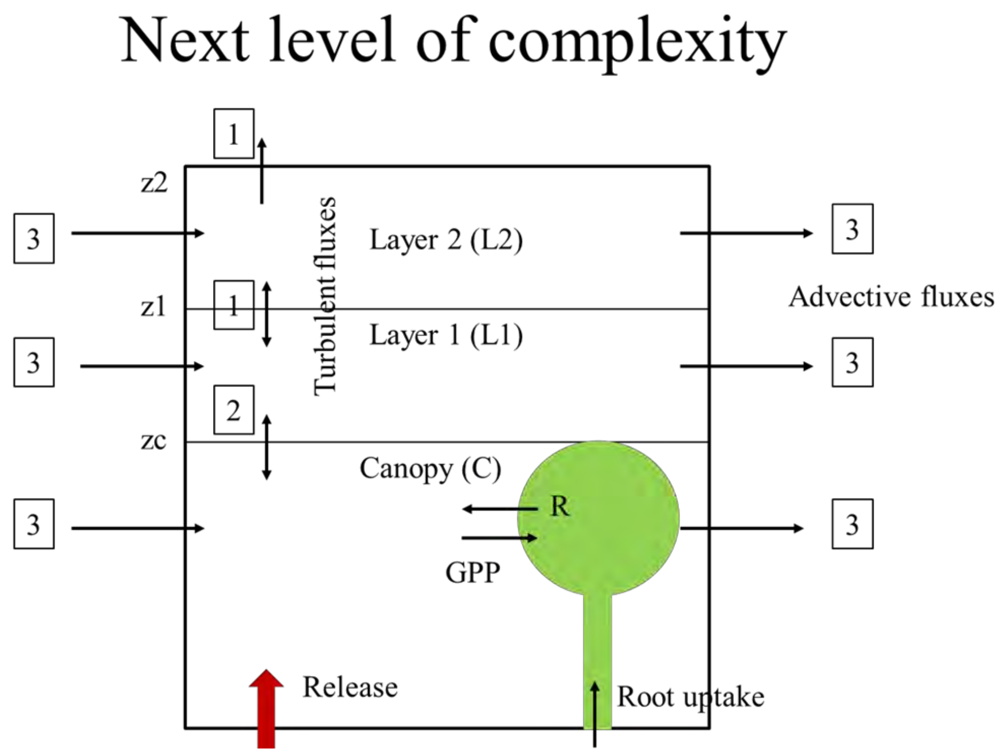

Figure 2-12. Facilia Model developed for SKB, Figure from SKB report in draft

The surface layer is divided into different vertical sub-layers: a canopy layer and two above-canopy layers (Layer 1 and Layer 2). The canopy layer might need be further sub-divided, especially for tall vegetation. For forest canopies it might be necessary to distinguish between an understory and a tree layer. More layers above the canopy could be added, but for the assessment purpose it is sufficient with only 2 layers. Layer 1 is needed if doses by inhalation are calculated. The lateral arrows (denoted with the textbox 3) represent advective fluxes. If the whole release area is represented with only one such model, then only the downstream advective fluxes directed from the biosphere object are considered. If the release area is divided into several adjacent sub-areas and a model is applied to each such sub-area, then the advective fluxes entering each sub-area from the adjacent sub-area are also considered. This allows modelling areas of difference size. The vertical arrows (denoted with textboxes 1 and 2) represent vertical turbulent fluxes. From Layer 2 there is a one-directional vertical flux out from the system (there is no back flux), which is equivalent to assume zero concentration of the contaminant (C-14) above this layer; as boundary condition. The GPP during day time and the respiration from above-ground parts of the plant during day time and night time should be given as inputs, on the shortest time scale possible. One possibility is to simulate a growing season on a daily basis, or a typical day on an hourly basis. Input of C-14 via respiration from the soil is assumed as being part of the release.

Root uptake is also an input to this model. It can be calculated from the Specific Activity in the soil solution, assuming a fractional contribution of root uptake to the NPP.

At steady state, hmix is not needed:

$$SA_c = \frac{\frac{Release\ rate}{Area}}{(1 - fra_{rootUptake}) * NPP + \frac{u_c * h_c * conc(C)_C}{\sqrt{Area}} + Flux_{C2L1} * conc(C)_C * RF_C}$$

The SA_c is the specific activity in the canopy atmosphere and in exposed primary producers. The first and second summands in the denominator have the same meaning as in the model used in previous assessments. The third summand represents the vertical flux of carbon between the canopy and above-canopy layers. The RF_c represents the fraction of carbon released from the canopy layer that is recycled back to this layer by vertical turbulence. The RF_c is a correction factor between zero and one that accounts for the fraction of C-14 that goes back to the canopy layer from the above-canopy layer. If RF_c is set to zero then all C-14 that reaches the above-canopy layer flows back to the canopy layer, which is equivalent to assuming that there is no flow from the canopy layer. In this case, the equation of SA used in Avila and Proel [2006] is obtained; if it is assumed that all C taken by the plant comes from the atmosphere. However, the parameters u and h have a different definition in the Avila and Proel [2006] model and in the model proposed here, where u is the average lateral wind speed in the canopy layer of height h_c. Setting RF_c to one is the same as assuming that there is no back flux of C-14 from the above-canopy layer to the canopy layer. This would lead to a slight underestimation of SA for cases with a large release area to a homogeneous field.

The concentration in air in a layer above the canopy of thickness h_{L1} can be used in estimations of inhalation doses to agricultural workers. For estimation of inhalation doses from staying at a forest area, the concentration in air in the canopy layer can be used. This can be calculated by multiplying the SA_c by the stable carbon concentration in the canopy layer.

The model was used to predict the wind speed profile below and above the canopy using data from forests, and eddy diffusivities with height based on the leaf area index (LAI). The model fits with observations in forests. The layer containing the minimum in the velocity profile acts as a block and changes with night and day and through the seasons. The average wind speed was used, and the specific activity was calculated for barley as shown in Figure 2-13.

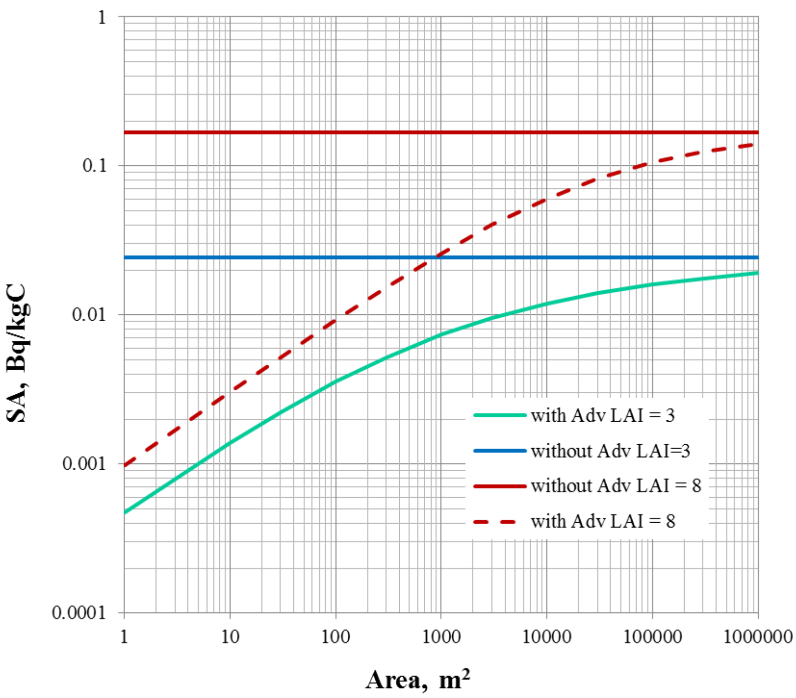

Figure 2-13. SA from 1Bq/m^2/y dependency on area, Figure from SKB report in draft

The horizontal lines represent the SA obtained if lateral advective fluxes are disregarded, corresponding to the values in the centre of a sufficiently large area where advective fluxes into and out of the area are identical.

Validation studies will compare values of RRI calculated with the model with values reported in the literature (C_{13}/C_{12} studies) and with values calculated with a Lagrangian model. Preliminary results calculate an RRI of 1.9%, consistent with a value of '<4.6%' (see Section 2.3). The Lagrangian model will be validated by comparing predicted CO_2 profiles with measured profiles. The model fits dense canopies well, however vegetables are only dense locally (e.g. when planted in rows or in gardens), so this may be more difficult to model.

2.6 TESTING ATMOSPHERIC-VEGETATION CARBON MODEL USING THE NORUNDA DATASET

Ivan Kovalets (UCEWP) presented.

The aim of the project, which was funded by SKB, was to test the parameterisation and validity of the C-14 assessment model developed by Facilia AB for SKB, to test the model's capability to predict the inside- and above- canopy profiles of CO_2 for different stability conditions, and to evaluate the parameters of the model. The project used data from the Norunda research station, established in 1994 as part of the NOPEX project for investigation of fluxes of momentum, heat, water, and CO_2 between soil, vegetation, and atmosphere in relation to climate change. Norunda research station is operated by Lund University and is located 30 km north of Uppsala in mature boreal forest, dominated by Norway spruce and Scots pine (H≈25 m, LAI=3 to 6). It has flat topography and contains a 102 m high measurement tower. Measurements of CO_2 and CH_4 concentrations, wind speed, temperature, humidity, are recorded from 8 to 102 m. Measurements of eddy flux, and micrometeorological data (heat flux, velocity fluctuations, etc) are recorded at a height of 35 m and the site has automated soil respiration chamber systems. Access to data with a 30 minute resolution is through the NBECC database http://dbnecc.nateko.lu.se/. The site participates in FLUXNET: a world-wide network of micrometeorological towers which use eddy covariance methods to measure the exchanges of CO_2, water vapour, and energy between terrestrial ecosystems and the atmosphere.

The presenter referred to the following publication: Lundin et al. [1998]. The general problem of atmospheric transport inside and above the canopy is illustrated in Figure 2-14.

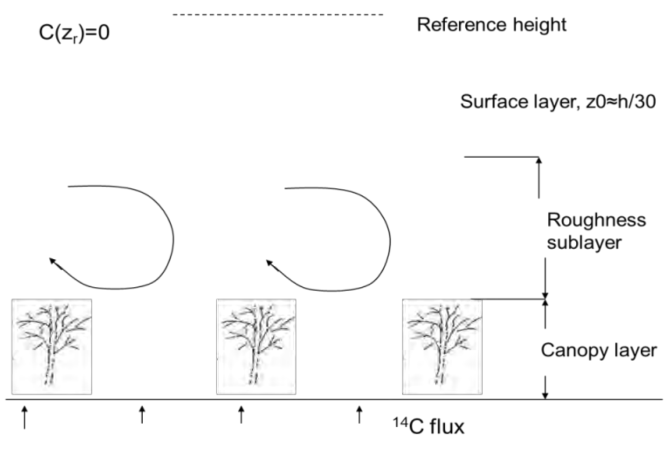

Figure 2-14. Atmospheric transport inside and above the canopy, Figure from SKB report in draft

Near the surface, but above the roughness elements, when shear production is dominant, the wind speed obeys a logarithmic wind profile:

$$U(z) = \frac{u_*}{k} ln\left(\frac{z-d}{z_0}\right)$$

where *k* is the von Karman constant, which has a value of about 0.4,
z_0 is the roughness length
u_* is the friction velocity.

The velocity profile and general modelling considerations are given in Figure 2-15.

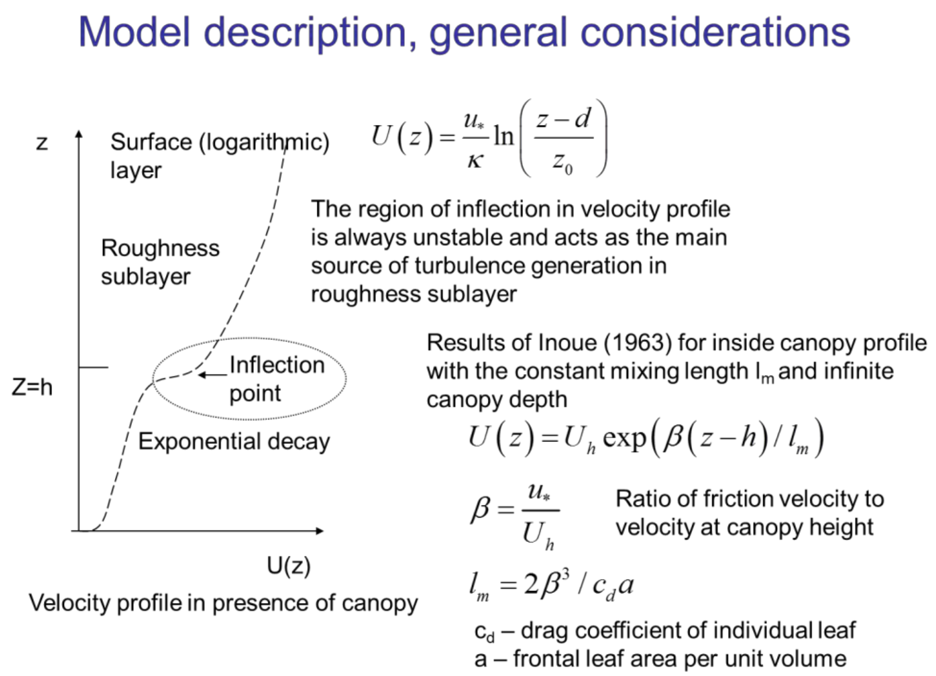

Figure 2-15. The velocity profile in the presence of the canopy and model description, Figure from SKB report in draft

The ratio of friction velocity to velocity at canopy height is expressed by $\beta=u^*/U_h\approx0.3$. This was obtained using direct measurements from Norunda station. A drag coefficient for the individual leaf (c_d) was obtained from literature (0.25). The leaf level Stanton number (r=0.1) was obtained from the literature. The frontal leaf area per unit volume (a≈0.14) was obtained by least squares fitting of the velocity profiles measured at Norunda.

Starting with the diffusion equation:

$$-d(K_c(dC/dz))/dz = S_x(z)$$

and considering the area above the canopy, roughness correction functions are of the form:

$$K_{m(c)} = \frac{\kappa u_*(z-d)}{\varphi_{m(c)}((z-d)/L)\hat{\varphi}_{m(c)}((z-d)/z_*)}$$

where Garratt [1980] has proposed:

$$\hat{\varphi}_{m(c)} = 1 - c_{m(c)}\exp\{-c_{2m(c)}(z-d)/l_{RSL}\}$$

and Harman and Finnigan [2008] have proposed:

$$l_{RSL} \sim U/(dU/dz)|_{z=h} = l_m/\beta$$

Within the canopy, this leads to:

$$K_c = l_m l_c (dU/dz) = \frac{2\beta^3 u_*}{a c_d S_{cc}} \exp\left(\frac{z-h}{2\beta^2 L_c}\right), z < h$$

where the turbulent Schmidt number inside the canopy layer $S_{cc}=K_m/K_c$ depends on the stability. Above canopy analytical relationships for coefficients $c_{m(c)}$ and $c_{2m(c)}$ entering the above relationships for the roughness sublayer correction functions $\hat{\varphi}_{m(c)}$ were derived in Harman and Finnigan [2008] by fitting the inside and above canopy profiles of concentration and momentum. Within the canopy, the simplified approach of Harman and Finnigan [2008] has drawbacks since the results are very sensitive to the value of β. Using Taylor's theory of turbulent diffusion [1921], an empirical parameterisation of the vertical velocity profile and Lagrangian timescale were obtained for the model.

$$K_c(z) = \sigma_w^2 T_L(z)$$

where:

$$\sigma_w(z) = \sigma_w(h)\left(\frac{z}{h}\right)^{\beta_1}$$

$$\sigma_w = c_\sigma u_*$$

The key parameters in the model, which control the above canopy mixing, are: the ratio of friction velocity to vertical velocity fluctuation at canopy height: $c_\sigma \approx 1.3$ (obtained using direct measurements from Norunda station); the rate of T_L decay with height: $c_1 \approx 0.4$ (obtained from literature); and the rate of vertical velocity fluctuation decay with height $\beta_1=1.5$ (obtained by fitting concentration profiles to Norunda data). This value of β_1 is consistent with β_1 values obtained from fitting published vertical profiles of σ_w, which range from 0.7 – 2.3.

The model was set up with a simulation period of 3 years (2007-2009), a time resolution of 30 minutes and a reference height (z_r) of 70 m. The source was assumed to be during the day time.

Stability ranges (approximately corresponding to the classifications of Golder [1972]) are defined by the Monin-Obukhov length L[a], where:

Neutral: L> 500 m

Slightly stable: 250<L<500

Moderately stable: 70<L<250

[a] L is defined to be negative in unstable conditions and positive in stable, and its magnitude is a measure of the height above which thermal effects become important, with shear production being dominant below |L|.

Slightly unstable: -500<L<-250

Moderately unstable: -250<L<-70

Respiration flux $R_F\,[\mu mol/m^2 s]$ from the soil surface was parameterized as a function of temperature [Lindroth et al. 1998]:

$$R_F = 1.64\,\exp(0.097t)$$

and the net photosynthesis flux (distributed through the upper part of canopy layer) was:

$$P_F = F_N - R_F$$

where the flux out of the top (F_N) was measured.

The concentration flux through the top of the canopy (c_*) was normalised using:

$$c_* = F_{top}/u_*$$

The results of the modelling were compared with measurements for different stability ranges and day or night conditions. The normalised average concentration results for day-time neutral stratification conditions are shown in Figure 2-16.

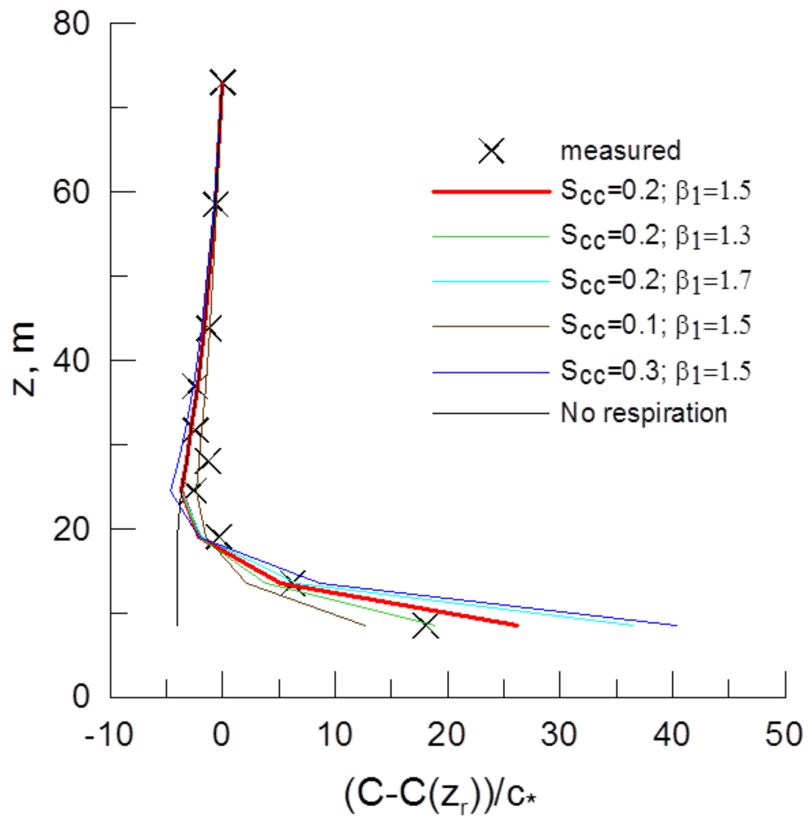

Figure 2-16. The velocity profile in the presence of the canopy and model description, Figure from SKB report in draft

It can be seen that the model fits the data well if respiration is included.

The model has been successful in reproducing vertical profiles of CO_2 concentrations for day-time and night-time conditions and for different stability conditions (from neutral to moderately stable and to moderately unstable). The successful results in reproducing the observed CO_2 profiles in day-time conditions could be achieved only by proper assignment of respiration flux. Taking into account the unstable stratification during day-time conditions reduces the inside canopy concentration by up to -25% as compared to neutral case. Thus, stability effects probably could be neglected in assessments of C-14 contamination. The results of an example simulation of C-14 in the Norunda area over 3 years are given in Figure 2-17.

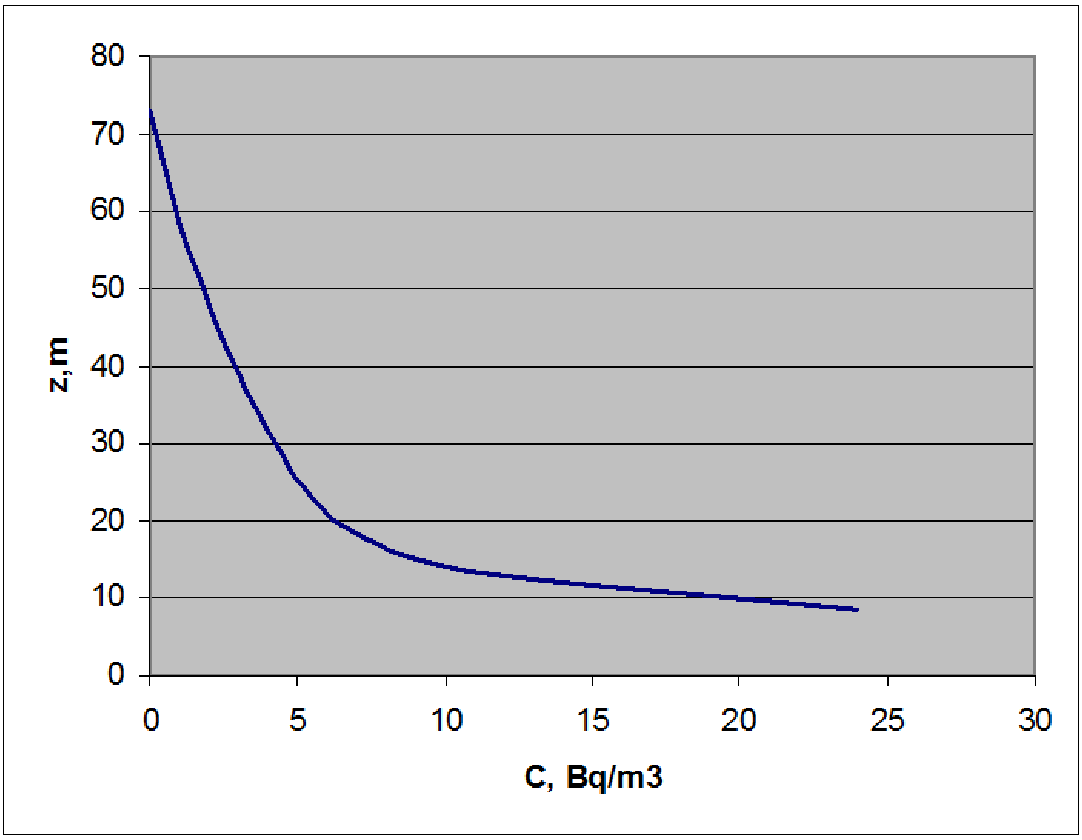

Figure 2-17. Average day-time concentrations of C-14 calculated for the conditions of Norunda over 3 years for the release of 1 Bq/m^2/s of C-14, Figure from SKB report in draft

Application of the model to other canopies may require some tuning of the parameters and additional studies are required in this respect.

2.7 FATE OF ACETIC ACID IN SOIL SOLUTION

Graham Smith presented on behalf of NIRS.

NIRS has a program of work for biosphere assessment for waste disposal. Several projects include work on C-13 and isotope ratios to determine carbon pools and fluxes to support understanding of C-14 behaviour. Three reports describing recent C-14 work by NIRS were summarised.

1) *Measurement of the fate of acetic acid form carbon in soil solution of flooded soils using high performance liquid chromatography coupled with isotope ratio mass spectrometry Keiko Tagami, Shigeo Uchida, Nobuyoshi Ishii GEODERMA Volume 165, No 1. 2011.*

C-13 labelled acetic acid was added to six soil–water mixture samples. The chemical form changes in the soil solutions were measured by high performance liquid chromatography coupled with isotope ratio mass spectrometry. After 24 hours, less than 3% of the carbon added as acetic acid remained in the soil solutions, and was possibly mainly in inorganic forms. Thus, for those soil types, most of the C-13 labelled acetic acid would likely be changed to other physico-chemical forms and be removed from the soil solutions in a very short time period. These results are consistent with Peter's results, presented earlier.

2) *Root Uptake of C-14 Leached from LLW for Sub-surface Disposal with Engineered Barriers {Rice} Suzuki, Ishii, Tagami and Uchida. Jpn J Health Physics 47(4) 2012.*

This study measured uptake of C-14 by paddy rice at different stages of plant development, studied using hydroponic conditions and acetic acid as the C-14 source. The plant root was soaked for two hours at different growth stages. Up to 2% of activity was recovered from the plant, with most activity in the root part of the plant at all stages. Mixing of root residue at the time of ploughing would result in enhancement of the persistence of C-14 in soil.

3) *Distribution coefficients of stable iodine in estuarine and coastal regions, and their relationship to salinity and organic carbon in sediments, Takata et al (2012). Environ Monit Assess.*

A number of assessments of Japanese coastal sites have shown that the distribution coefficient (Kd) of stable iodine decreases along the salinity gradient, from river to sea. However, at the higher end of salinity range, there is still a wide range of variability in Kd. At most of the sites investigated, this is correlated with the levels of organic carbon.

2.8 DISCUSSION POINTS

In addition to clarifying points in the presentations, the discussions explored the further use of the IRSN dataset, for example whether it would be possible to examine air measurements on days where the release was low to identify soil respiration. The potential of the NDA experimental work was also discussed. In long-term assessments, accumulation in soil should be considered as the land use and crops may change over time and the C-14 may persist in the soil after the release has ceased.

3. CARBON-14 IN AQUATIC ECOSYSTEMS

The focus of BIOPROTA on issues relating to C-14 has, to date, been terrestrial systems, specifically release from soils, behaviour in the plant-canopy atmosphere and uptake into plants. However, C-14 is also an issue for post-closure assessments for radioactive waste disposal in aquatic ecosystems. As such, a review of the behaviour of C-14 in aquatic systems has been undertaken, including consideration of approaches that have been taken to model transfers between environmental media (sediment, water, air) and biota. The key findings of the review are presented below. These provided the basis of an introductory presentation on C-14 in aquatic systems by Karen Smith (RadEcol Consulting). Presentations provided by workshop participants relating to C-14 in aquatic environments are also summarised, and discussion points arising from all aquatic system presentations are summarised in section 3.4.

3.1 REVIEW OF C-14 BEHAVIOUR IN AQUATIC ECOSYSTEMS AND APPROACHES TO MODELLING

Carbon-14, transported from the geosphere by advection with groundwater is likely to discharge to low-lying areas of the landscape, which will often coincide with surface water bodies such as wetlands, lakes or coastal waters. Surface water bodies may therefore be directly impacted by the release of C-14, with the potential for uptake by plants and animals. Indirect transport to water bodies may also occur following extraction of groundwater for agricultural processes. This may include atmospheric transport with transport across the interface between air and water and/or lateral transport in soil water to surface water bodies.

Once within the water, C-14 may be incorporated into biota through photosynthesis of free CO_2 or bound CO_2 (carbonate/bicarbonate) or by consumption of particulate organic carbon (POC). Processes for assimilation of C-14 into biota depend upon the form of the water body (depth, stratification etc.) and water flow conditions.

An experimental study on the behaviour of C-14 in freshwater systems was performed on a Canadian Shield lake [Stephenson et al. 1994]. The primary objectives were to measure rates of primary production and to study processes of gaseous exchange with the atmosphere [Stephenson et al. 1994]. These aspects were studied during the initial stages of the field study with subsequent evaluation of the fate of C-14 in sediments, water and biota around 10 years following initial addition to the lakes [Stephenson et al. 1994]. The results of the experiment have been used as the basis for a model inter-comparison exercise performed within the BIOMOVS II programme [BIOMOVS II 1996; Bird et al. 1999] and for model development (see section 3.2.3).

The experimental area comprised three lakes: L224 and L226N and L226S.

Lake 226 is essentially a double-basin lake with a surface area of 16.1 ha, with a shallow channel dividing the lake into two basins of approximately equal area (8.3ha in the north and 7.8 ha in the south), referred to as L226N and L226S [Stephenson et al. 1994; Bird et al. 1999]. A single outflow is located in L226N; no permanent inflows are present. L226N and L226S are connected by a narrow channel allowing mixing of water between the two basins. Four additions of C-14 labelled bicarbonate (dissolved inorganic carbon, DIC) were added to L226N in 1978 and a single addition was made to L226S. Each individual input was 11.5 GBq.

Lake 224 (L224) is a deep oligotrophic basin with a surface area of 25.4 ha and a maximum depth of around 27 m [Stephenson et al. 1994]. L224 received a single input of 37 GBq of C-14.

3.1.1 Cycling of C-14 aquatic systems: results from field experiments

Following addition, the pool of C-14 labelled DIC throughout L226 was rapidly depleted by both gaseous evasion and photosynthetic uptake. Of the C-14 added to L226S, 18% was lost to degassing and 12% to epilimnetic sediments, the outflow and deeper waters, whereas for L226N which received four consecutive inputs of C-14 labelled DIC, 26% was estimated to be lost to degassing and 22% to epilimnetic sediments, the outflow and deeper waters. In L226S, losses occurred in the first 22 days. However, in L226N the bulk of C-14 loss from degassing occurred in the first 10 days: increased lake productivity after 10 days served to increase pH (>pH 9) which served to limit gaseous evasion. It was estimated that the rate of evasion in L226S was 9 $mmol/m^2/d$ in the first 12 days, dropping to half this rate for days 13 to 29 compared with 0 to 25 $mmol/m^2/d$ in L226N [Bower, 1981, cited in BIOMOVS II, 1996]. Overall a transfer rate from water to atmosphere in L226S of 2.5 to 5.5 per year was calculated and 8 to 21 per year whilst degassing occurred in L226N. Of the C-14 remaining in L226N and L226S in the initial days following addition, the majority was fixed through photosynthetic processes resulting in particulate organic carbon (POC). Whilst the majority of gaseous evasion was observed in the initial days following addition, the atmospheric pathway for loss of organic carbon is likely to remain important in terms of the metabolism of DOC and POC to CO_2, which may be re-assimilated by primary producers or lost to atmosphere.

In L224, rapid mixing throughout the epilimnion was observed following addition of C-14. Subsequently however, most of the C-14 was lost via CO_2 exchange with the atmosphere; a smaller fraction was again fixed through photosynthetic processes resulting in POC [Stephenson et al. 1994].

Fiévet et al. [2006] measured DIC in seawater at various points along the French shoreline affected by discharges of C-14 from the AREVA spent fuel reprocessing plant at La Hague in France. Results indicated that some loss to atmosphere (or other sink) may occur, but that this loss was not significant as compared with data scatter.

Whilst gaseous evasion of CO_2 was noted as an important loss mechanism for the experimental lakes, invasion from atmosphere may also occur [Bontes et al. 2005]. This is particularly the case where DIC in the water column is depleted, for example due to sedimentation of POC in stratified systems or through incorporation in low carbon turnover flora, such as in systems dominated by macrophytes as opposed to phytoplankton primary production and where heterotrophy is low (i.e. reduced respiration). Again, this points to food webs being important in governing the carbon cycle in aquatic systems – the floral community, which itself governs the faunal community, is important in determining the rate of carbon turnover.

In the studies on Canadian Shield lakes, following the rapid depletion of the DIC pool, the C-14 inventory was controlled by the sedimentation of POC. Thus, despite the importance of gaseous evasion to the atmosphere to the short-term fate of C-14 in these lake ecosystems, the lakes retained significant quantities of C-14 in sediments. For example, in year 11, 16.9% of the initial inventory of C-14 remained in the sediments in L226N with a peak of activity being observed in the sediment profile at a depth of around 4 cm, indicating a low post-depositional mobility for sediment particles incorporating C-14 [Stephenson et al. 1994]. The high retention of C-14 in sediments observed in L226N may, in part, have arisen from the increased productivity observed in the initial days following addition. This served to reduce gaseous evasion which may have led to a greater fixation of C-14 into phytoplankton which in turn may have increased the rate of sedimentation of POC and loss to sediments. The peak of C-14 activity in L226N sediments was not observed in all cores from the experimental lakes and appeared to be affected by the degree of bioturbation: oxic regions, consistent with high sediment dwelling invertebrate populations, led to C-14 being more uniformly distributed.

The decomposition and re-suspension of C-14 in sediments continued to provide fluxes of DIC, DOC (dissolved organic carbon) and resuspended POC to the water column over time and continued to provide a source of carbon to lake-dwelling organisms. C-14 concentrations in DIC were higher at depth than in surface waters with the maximum activity concentration being recorded at the sediment-water interface. In addition to sediments providing a continued source of C-14 to the lower water column, differences observed in C-14 concentrations throughout the water column may also, in part, have resulted from isotopic exchange of C-14 with stable carbon in the atmosphere, thus leading to C-14 depletion in the surface waters. Stratification of lakes may tend to enhance these differences, resulting in benthic organisms having greater exposure to, and therefore greater potential for assimilation, of C-14 than pelagic species. The mean specific activity of the DOC pool of C-14 in lake water was between 15% and 19% of the specific activity of the DIC pool [Stephenson et al. 1994]. The cycling of C-14 was evident more than a decade after initial addition of C-14.

In terms of biota within the experimental lakes, activity concentrations were measured in whitefish with the peak activity concentrations occurring in year 1. Smaller fish had the highest concentrations, indicating that juveniles were actively growing and thus depositing carbon with a high specific activity of C-14 in the tissues generated during this active growth period [Stephenson et al. 1994]. Over time larger fish had higher C-14 concentrations than small (new recruit) fish. The observed lag between the addition of C-14 to the lake and the peak specific activity of C-14 in fish tissue suggested a relatively slow rate of metabolic tissue replacement at around 1 per year [Stephenson et al. 1994]. Observations of high levels of C-14 in fish more than a decade after its addition illustrates that C-14 remained bioavailable and important within the food chain [Bird et al. 1999]. Consistent with the peak activity concentration in whitefish, radiological risk to people was dominated by the fish consumption pathway and peaked around 1 year after addition of C-14.

Samples of other biota within the experimental lakes indicated that plants were in equilibrium with the surface water DIC pool, invertebrates showed varying degrees of dependence on algal (i.e. DIC based) carbon and sediment carbon as did various fish species [Stephenson et al. 1994]. DIC has also been found to be the primary source of carbon for primary producers and is considered the most representative of the carbon pools in marine systems upon which to calculate concentration ratios between seawater and brown algae [Douville et al. 2004, cited in Fiévet et al. 2006]. Fiévet et al. [2006] also investigated the transfer of different forms of C-14 to a range of marine species (macro-algae (*Fucus sp.*), molluscs, crustaceans and fish) collected at a variety of distances from the point of liquid effluent discharge from the AREVA spent fuel reprocessing plant at La Hague in France. Results indicated that peak concentrations in DIC were not mirrored by biota suggesting that transfer kinetics smooth out variations in seawater DIC [Fiévet et al. 2006]. However, it was noted that the input of C-14 to biota from sources of C-14 other than DIC was not investigated, which leads to uncertainty, particularly for non-photosynthetic species within a food web. Indeed such differences were observed by Stephenson et al. [1994] and Cook et al. [1998, 2004, as cited in Fiévet et al. 2006].

The food web is therefore important in governing the uptake of C-14 in the aquatic environment. From the experiments it can be concluded that biota dependent on freshly fixed algal carbon have relatively low exposure to C-14 due to rapid depletion of DIC[a] whereas biota dependent on detrital carbon sources are exposed to a much greater degree and are not in equilibrium with the specific activity of DIC in the water column. The residence time of C-14 in fish tissue is such that contamination of fish may persist long after the specific activity of C-14 in the DIC pool has declined. Indeed, Sheppard et al. [2006], on the basis of results from Stephenson et al. [1994], calculated a turnover rate in whitefish

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[a] It is important to note that this rapid depletion and knock-on effects for uptake in the food chain relates to distinct point source releases and not continued inputs as may be expected following release from a geological disposal facility for radioactive waste.

of at least 0.17/y, based on observations that specific activities in fish were much higher than that in the food items at the time of sampling, implying that fish tissue was reflecting specific activities of food items from several years previous.

Of the carbon sources in an aquatic environment, zooplankton (pelagic primary consumers) have been demonstrated to preferentially select phytoplankton as their principal food source, irrespective of the availability of other POC, including that from terrestrial sources [Zigah et al. 2012].

3.1.2 Cycling of C-14 in aquatic systems: results from modelling studies

In 2001, SKB performed a modelling assessment to evaluate the behaviour of C-14 in brackish water following a hypothetical release from the repository for operational radioactive waste, SFR-1, into a bay of the Baltic Sea [Kumblad 2001]. The model was based on an ecosystem approach whereby general ecological principles identifying and quantifying the main flows and storage of energy (carbon) and C-14, both in the physical environment and the food web was employed. The energy flows were based on quantified carbon budgets and fluxes measured in the region to which the model was applied. For the model simulation, it was assumed that C-14 (~5E7 Bq/y over 1000 years, corresponding to the estimated maximum discharge) is released as inorganic carbon (CO_2 or carbonate ions) from the repository to the brackish water system and is thus available to autotrophic organisms in both pelagic and benthic environments. Uptake of C-14 to fish was based on data on the fish species distribution in the area and their feeding habits. A subsequent revision of the model [Kumblad and Kautsky, 2004] was also applied to evaluate the behaviour of C-14 following a hypothetical release to the Bay. This model was similar in structure, but incorporated nutrient cycling as a means of taking account of an over-simulation of primary production. Further detail on the models is provided in section 3.1.3.

Consistent with the results from the Canadian Shield lake experiments, the greatest uptake from model simulations is to benthic organisms, irrespective of whether the release is assumed to be direct to the DIC pool (homogenous distribution) or to the benthic compartment. However, whereas gaseous evasion and sedimentation of POC were primary factors in defining the C-14 distribution in the lake experiments, enhanced accumulation in the benthic food web relative to the pelagic in the model simulation was accounted for by the high water exchange within the bay, leading to the flushing of plankton, combined with a high biomass of benthic primary producers that are less affected by water flushing. This again points to the benthic community as being of importance for C-14 modelling in the aquatic environment, but also identifies the rate of water exchange as an important consideration. Lower water exchange with the Baltic Sea, simulated for a release occurring in 4000 AD when mixing between brackish waters within the Bay and the outer Baltic Sea will be reduced, led both to equilibration times being extended and overall uptake of C-14 is increased due to retention within the bioavailable pools, i.e. ecological half-lives are increased [Kumblad, 2001; Kumblad and Kautsky, 2004]. Unlike the Shield Lake experimental results, gaseous evasion was a minor loss mechanism, accounting for only 0.02% of C-14 losses under high water flow conditions, increasing to 1.5% under reduced flow conditions at 4000 AD. However, in a further developed model applied to a similar release scenario, it was estimated that 9% of the annual primary production is exchanged over the air-sea interface as respired CO_2 [Kumblad and Kautsky, 2004].

According to model results [Kumblad, 2001], of the C-14 assimilated into biota, around 4% was then recycled back to the water column as a result of respiration under high water exchange conditions, increasing to 21% under reduced flow conditions. Loss from organism compartments (death, gamete production etc.) was the key biological route for C-14 flow in the system, with >90% of the POC compartment being exported annually from the system via water exchange. The majority of the remaining POC was consumed by benthos with only 0.25% being buried in sediments each year (increased under low flow conditions).

A wide range of bioconcentration factors were calculated based on the results of the simulations [see Table 6-4, p. 44 in Kumblad, 2001], which vary at different points in the simulation indicating the importance of ensuring steady state if applying concentration factors as a means of deriving biota concentrations following a release to the biosphere.

3.1.3 Modelling approaches for C-14 in aquatic systems

A number of models have been developed to represent C-14 behaviour in aquatic systems. These vary in their approach to representing C-14 behaviour and their level of complexity. Conventional models, based on transfers of C-14 between different environmental and biota compartments were applied in a model inter-comparison exercise that was conducted within the BIOMOVS II programme, which ran from 1991 to 1996, and was an international cooperative programme aimed at evaluating models for the transfer and accumulation of radionuclides and other trace substances in the environment. Additional models, not applied within the BIOMOVS II exercise are also described in the following sections.

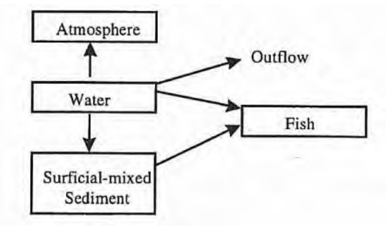
Models used for regulatory decision making are generally no more complex than those applied within the BIOMOVS II C-14 exercise. However, models developed for waste disposal assessment have included more complex representations of C-14 behaviour, based around specific activity and carbon budgets and fluxes, as illustrated by the SKB [Kumblad, 2001; Kumblad and Kautsky, 2004] model.

BIOMOVS II model inter-comparison

Four models were employed in a C-14 model validation and comparison exercise within the BIOMOVS II programme. The C-14 exercise, reported in BIOMOVS II [1996] and Bird et al. [1999], was based around the Canadian Shield Lake experiments discussed in section 3.1.1. The primary objectives of the exercise were:

- To test the accuracy of the predictions of environmental assessment models for selected contaminants and exposure scenarios;
- To explain differences in model predictions due to differences in model structure, modelling assumptions and/or differences in selected input data; and
- To recommend priorities for future research to improve the accuracy of model predictions.

Scenarios were developed on the basis of data derived from the Canadian Shield Lake experiments and used as a basis for simulating the fate of C-14 added to the lakes. Models were therefore applied to predict C-14 concentrations in lake water, sediment and whitefish over a 13-year period. The four models applied are illustrated in Figure 3-1.

AECL Model 	• Simple mass balance model of a lake • Doesn't explicitly account for release of C-14 from sediments back to water, hence water predictions less accurate than for the more complex models, however sediment concentrations were closest to observed values due to degassing losses exceeding rate of loss to sediments • Predicted rapid loss of C-14 from water • Fish concentration predicted on basis of specific activity in water immediately following C-14 addition and in sediments at year 13.

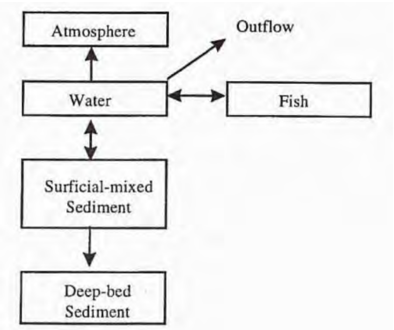
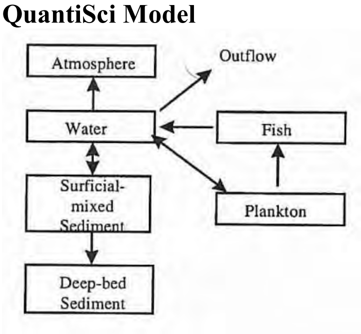
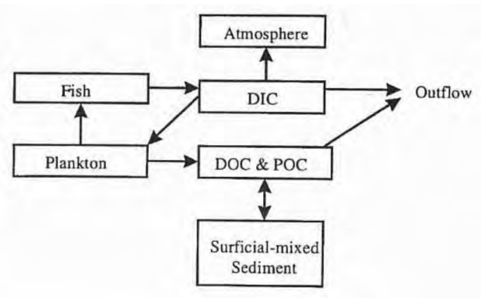
<table>
<tr>
<td>Studsvik Model A

</td>
<td>
Complex deterministic model
Less rapid loss of C-14 inventory from water which is never completely depleted (cycling between sediment and water considered)
C-14 release from sediments; some recycling back to sediments, but greater loss from gaseous evation (as compared with QuantiSci model)
</td>
</tr>
<tr>
<td>QuantiSci Model

</td>
<td>
Relatively complex deterministic dynamic compartmental model
Considers recycling within the lake
Less rapid loss of C-14 inventory from water which is never completely depleted (due to cycling between water and both sediments and plankton)
C-14 release from sediments; most recycled back to sediments
Dynamic uptake of C-14 into fish: uptake by fish into flesh is only small proportion of the carbon consumed with remainder being excreted. Uptake factor of 1% assumed.
Overestimated C-14 retention in lake sediments
</td>
</tr>
<tr>
<td>Studsvik Model B

</td>
<td>
Complex probabilistic model
Considers recycling within the lake
Gives specific consideration to DIC, DOC and POC
Less rapid loss of C-14 inventory from water which is never completely depleted
Most accurate in simulating C-14 in water and whitefish
Overestimated C-14 retention in lake sediments
</td>
</tr>
</table>

Figure 3-1. Models applied in the BIOMOVS II C-14 exercise and key processes and differentiators between models.

Each model produced reasonable predictions when compared with observed data and when uncertainty was taken into consideration (BIOMOVS II, 1996). Experimental data indicated that around 0.2 to 0.4 % of the initial C-14 inventory added to the lakes remained in the water column at the end of the study due to recycling of C-14 from sediments. The simple AECL model did not account for this recycling of C-14 and, as such, predictions were not as realistic as with the other deterministic and probabilistic models in terms of C-14 concentrations in water. However, sediment concentrations derived using the AECL model were closest to the observed values. Sediment concentrations were more difficult to predict due to differences in sedimentation rates assumed in models and assumptions around the amount of uncontaminated sediment that the C-14 mixes with. The predicted C-14 inventory in sediments was therefore considered a better endpoint [Bird et al. 1999]. Models were found to be conservatively high for the C-14 inventory in sediments (within a factor of 2-11 of observed values) with uncertainties largely being associated with the transfer rate from water to sediment and gaseous evasion.

All models predicted a rapid uptake of C-14 into fish with a then gradual decline throughout the simulation period. Studsvik model B provided the most accurate simulation of water and whitefish concentrations, but over-estimated sediment concentrations. The QuantiSci model also overestimated C-14 retention in sediments. All models under-predicted fish concentrations in the scenario developed around L226N for which high productivity was observed in the days following C-14 addition, which served to retain C-14 within the lake basin due to reduced gaseous evasion.

Based on the observed continued bioavailability of C-14 following input, Bird et al. [1999] concluded that relatively complex models with internal lake recycling of contaminants were required for modelling C-14 over the long term.

The AECL dynamic C-14 model

Whilst the AECL C-14 model applied within BIOMOVS II was a simple mass balance approach, a more dynamic model has been developed [Stephenson and Reid, 1996] and calibrated using the data collected from L226N and validated through application to L226S and L224; hence it was not appropriate to apply within the BIOMOVS II programme. The model takes into account seasonal variations in the physical, chemical and biological processes of carbon transport in lakes and was thus an enhancement to the model applied within BIOMOVS II.

The dynamic C-14 model takes into account the activity of C-14 in a number of aqueous, sediment and food-chain compartments including the epilimnion and hypolimnion; DIC, DOC, POC; surface and buried sediments in the erosional and depositional zones and losses to an outflowing stream and to the atmosphere. DIC, DOC and POC pools are distinguished between those in the epilimnion and those in the hypolimnion and these two zones are considered to behave separately with the exception of two brief mixing periods per year (spring and autumn) when the water column is assumed to be thoroughly mixed: the inventories of C-14 in the two pools are summed and re-apportioned according to the relative volumes of the epilimnion and hypolimnion. Ice coverage with season is also taken into account; when ice is covering the lake, no loss to atmosphere is assumed to occur. The water and sediment compartments within the model are assumed to process carbon at different rates, reflecting known physical, chemical and biological processes. Water of the epilimnion can leave via an outflowing stream with DIC, DOC and POC also discharging at rates proportional to the water flow, which is variable with season.

DIC in the water may be lost to atmosphere (excluding during ice cover) or fixed as POC by phytoplankton. POC can sink through the water column throughout the year except for periods of lake water mixing. The DOC pools are supported by release of decomposition products from POC and sediments or by excretion from organisms. DOC is decomposed to DIC or used as a carbon source by organisms comprising the POC (i.e. microbes).

Sediments that are in contact with the epilimnion are assumed to be erosional whereas those beneath the hypolimnetic water column are assumed to be depositional. Erosion zone sediments are subject to resuspension throughout the summer whereas depositional zone surface sediments are subject to resuspension twice each year; one day prior to mixing of the epilimnion with the hypolimnion. Decomposition rates of depositional zone sediments do not vary seasonally since water temperatures do not vary much from 4 ºC.

As compared with the simple AECL model applied within BIOMOVS II, the dynamic C-14 model was shown to be much better at predicting C-14 behaviour. However, over the long-term the model predictions converged since the former is specifically designed to simulate long-term net exchange between water and sediment assuming a continuing source-term flux. Therefore the simpler model was considered adequate for predicting the fate and concentrations of C-14 in lakes given loadings that vary only slowly, allowing chemical conditions to approach steady state [Stephenson and Reid, 1996].

Modelling long-term release of C-14 from waste disposal facilities: the SKB approach

As noted in section 3.1.2, the SKB modelling approach [Kumblad, 2001; Kumblad and Kautsky, 2004] is based around transfer of mass and energy (i.e. energy-based systems ecological modelling) where carbon transfers are based on quantified carbon budgets and fluxes in the ecosystem. The approach is based around two model sections: one for carbon flow based on results the carbon budget, and the other for C-14 flow. The model structure is illustrated in Figure 3-2. The C-14 section differs from that illustrated by incorporating an additional compartment (SFR C-14) for the input of C-14 to the system following repository release. Together, the carbon and C-14 sections provide the basis for calculation of exposure.

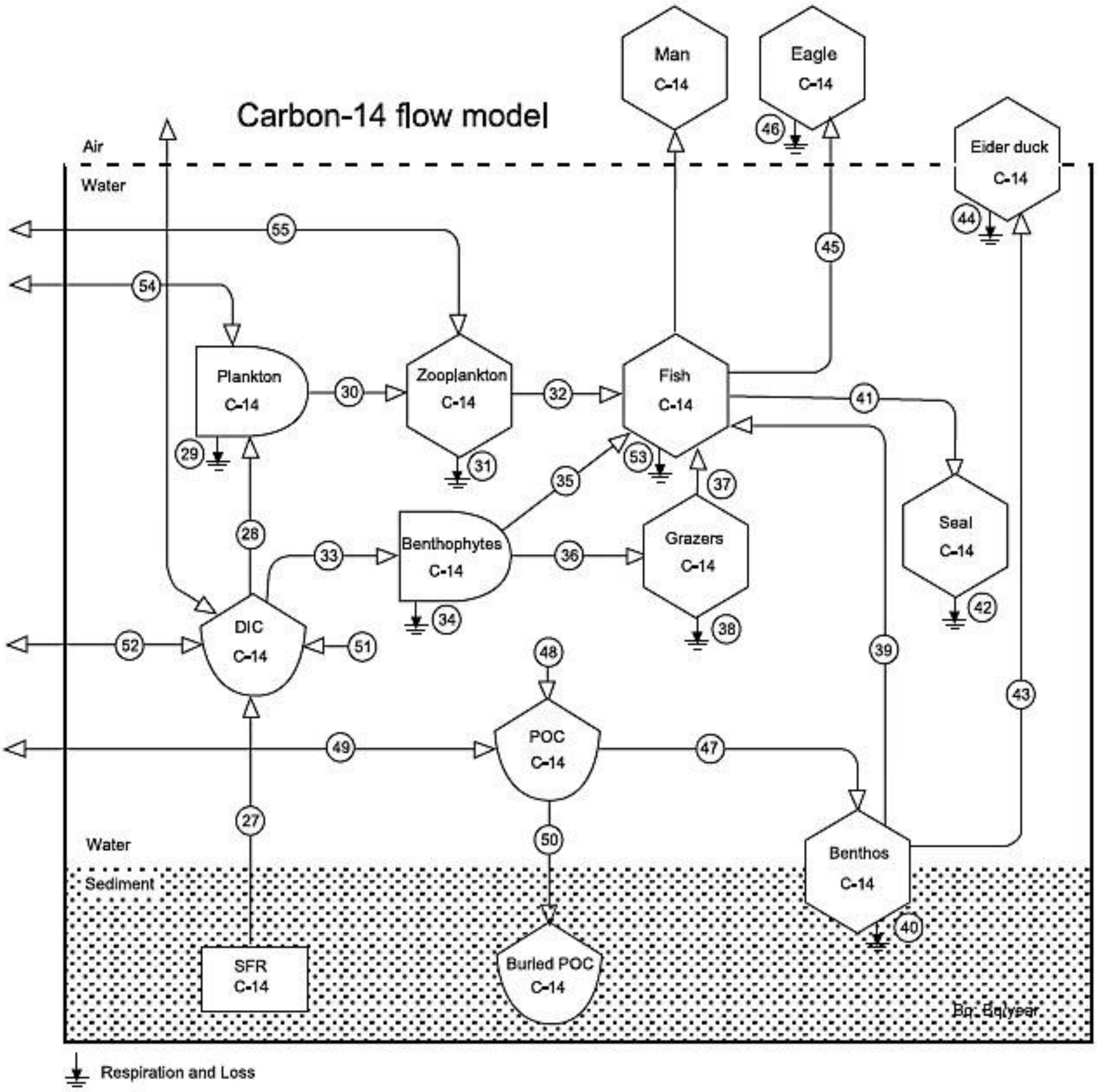


Figure 3-2. The SKB model for C-14 in brackish environments [Figure 4-2 from Kumblad, 2001].

In the revised model [Kumblad and Kautsky, 2004], the carbon component has been revised to incorporate the cycling of other nutrients (nitrogen and phosphorus), giving a CNP model which is again linked to the C-14 model. The revision was triggered by the realisation that DIC was a limiting factor in the original [Kumblad, 2001] model, which was interpreted as arising from an over-estimation of primary production. Additional nutrient dynamics were therefore incorporated to improve estimates of primary productivity. C-14 flow continued to be modelled in the revised model [Kumblad and Kautsky, 2004] in proportion to the presence of stable carbon, i.e. an isotopic approach. C-14 entering the system through the compartment 'SFR C-14' is assumed to be available both to pelagic and benthic primary producers via the DIC-14 pool.

Since primary production is to a great extent dependent on solar radiation, seasonal variations in production are compensated for by normalising the primary production per light-day (a day with a minimum insolation of 5 MJ/m^2) and multiplying with the annual number of light-days. Seasonal changes in respiration rates are also compensated for by considering temperature variations throughout the year. Consumption of food web organisms by fish was modelled according to the distribution of the fish species in the area and their feeding habits. Loss from all biota compartments is collected in a 'total loss' flow variable which feeds the POC compartment for which the carbon content varies according to the exchange of POC with that in the outer bay.

Modelling long-term release of C-14 from waste disposal facilities: the Posiva approach

Consistent with the approach employed by SKB, Posiva has also adopted a specific activity approach to the modelling of C-14 in aquatic systems. The model, described in Avila and Pröhl [2007] is illustrated in Figure 3-3.

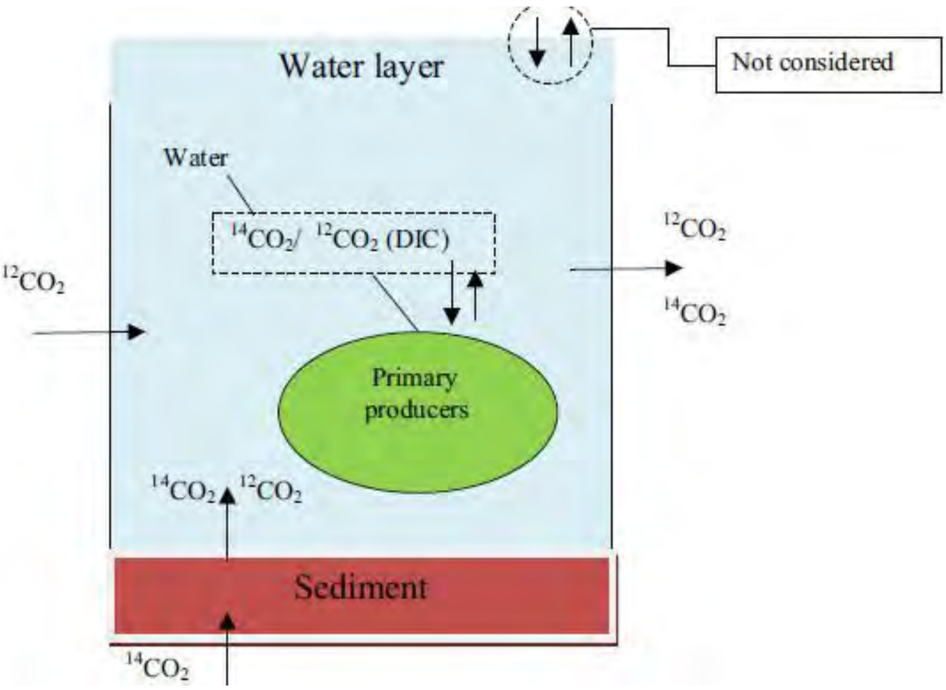


Figure 3-3. Posiva model for C-14 in aquatic environments [Figure 4-1 from Avila & Pröhl, 2007].

The model assumes that, C-14 released from a repository in the form of CO_2, enters the bottom sediment of a lake or sea basin where it then reaches the water column and mixes fully with stable inorganic carbon (DIC). The DIC enters the food chain by assimilation by photosynthetic primary production. Whilst it is acknowledged that assimilation of C-14 will occur only during the period of active photosynthesis, it is conservatively assumed that 100% of the release to the system occurs when photosynthesis is active [Avila and Pröhl, 2007].

The specific activity of C-14 established in the water column[a] is conveyed along the food chain to man. No loss by gaseous evasion is assumed in order to maintain conservatism in the model.

3.1.4 Key Aspects in the Behaviour of C-14 in Aquatic Systems

Based on the above review of C-14 behaviour in aquatic systems from field experiments and modelling studies, the following key considerations were presented during the workshop:

- Gaseous evasion is a potentially important loss mechanism for C-14 from water bodies; however the significance of the rate of release is affected by factors including water exchange rate, biological productivity and the surface area of the water body. Gaseous evasion will also be restricted during periods of ice cover although release may then occur when the ice recedes.

- DIC is assimilated in primary producers. In water bodies where phytoplankton dominate, POC can be sedimented thus providing a source of C-14 to sediments. However, POC sedimentation will be greatly reduced in systems with high rates of water exchange. Under such water exchange regimes, assimilation by macrophytes is likely to be the important route for C-14 to enter the food chain, particularly in shallow water and/or clear water bodies with high light penetration. Depth of a water body is therefore also an important consideration since this will affect the biomass of benthic primary producers and the benthic food chain.

- Increased productivity affects both pH, which reduces degassing under alkaline conditions, and increases fixation in algae which in turn may increase loss to sediments.

- Sediments play a key role in the cycling of organic carbon. POC can settle to sediments and provide a source of DOC/DIC and resuspended POC to the overlying water.

- C-14 concentrations in biota appear to be greater for benthic as opposed to pelagic organisms. This is particularly the case in stratified water bodies where sedimentation of POC and cycling between sediments and the hypolimnion results in greater activity concentrations being available to biota than in the epilimnion. This difference may be enhanced due to isotopic exchange across the air-water interface. The difference in C-14 uptake between pelagic and benthic organisms is also evident in high water exchange systems, as evidenced by the model simulations performed by Kumblad [2001] and Kumblad and Kautsky [2004].

3.2 CARBON-14 DYNAMICS IN A WETLAND ECOSYSTEM (DUKE SWAMP)

Tamara Yankovich (Saskatchewan Research Council) presented.

Wetlands are of considerable interest internationally, providing an interface between terrestrial and aquatic ecosystems. They are largely marsh, fen, peatland or water areas with a relatively shallow water depth (< 6 m). They occur at the interface between the terrestrial environment and both coastal and freshwater systems and are largely considered to be both biologically productive and diverse systems. Many wetlands fall under international protection treaties, e.g. the RAMSAR Convention. Historically however, many wetlands have been affected by discharges of radioactivity from sources including the nuclear industry and research laboratories. This is the situation with Duke Swamp, which is a 0.102 km^2 wetland located on AECL's Chalk River Laboratories (CRL) site in Canada. The swamp has received historical inputs of radionuclides, including C-14 and H-3, from hospital waste

[a] The specific activity established in the water column relates to the C-14 entering the system from the repository; the presence of natural C-14 is not taken into account.

that is stored upstream in a waste management area. Assessments on the exposure of plants and animals in the region have indicated that doses are dominated by the contribution from C-14, which has triggered field surveys to investigate how C-14 is distributed spatially throughout the wetland and whether biota concentrations reflect those calculated on the basis of a specific activity model for uptake by plants and animals [Yankovich et al., 2008a, 2008b, 2013].

A comprehensive vegetation study has been undertaken at 69 locations across the site and activity concentrations compared with those in a groundwater upwelling area where previous measurements had been made to determine the spatial scale of impact and how activity concentrations have changed over time [Yankovich et al., 2008a]. The spatial extent of contamination indicates that contamination is primarily localised within 0.1% of the swamp surface area [Yankovich, 2008a, Yankovich et al, 2013], see Figure 3-4.

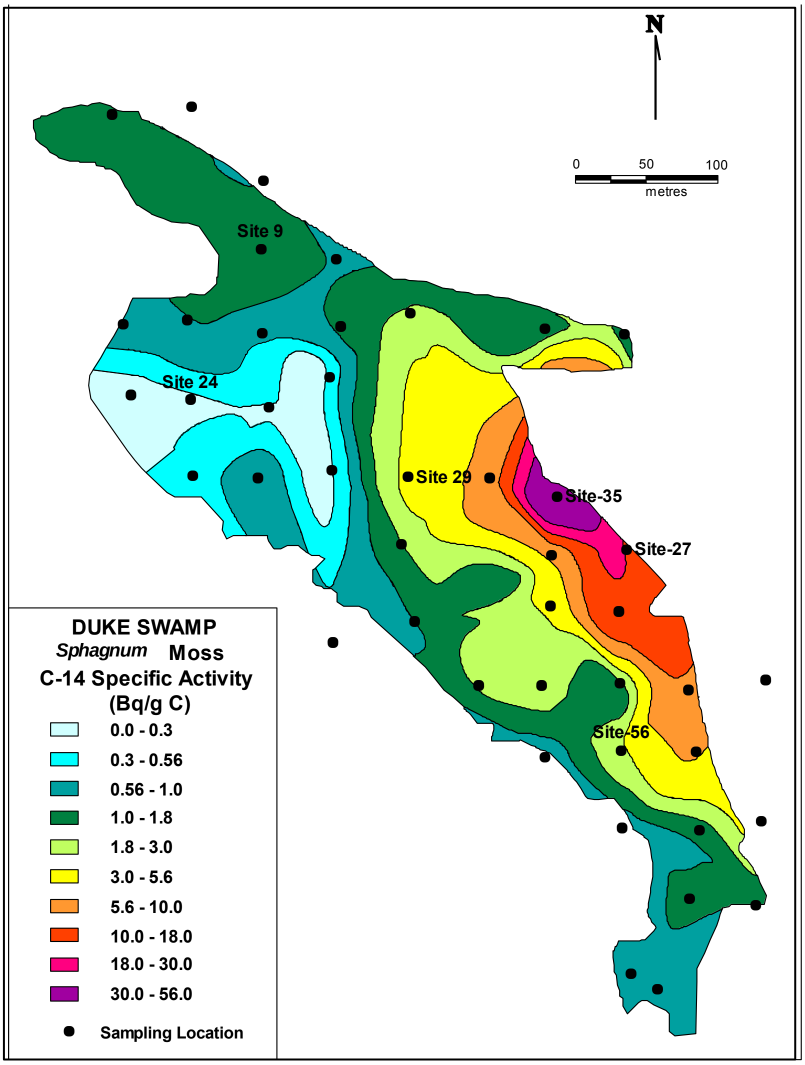


Figure 3-4. Spatial distribution of C-14 in vegetation samples (sphagnum moss) at Duke Swamp [Yankovich et al., 2008a]

Previously, the area with higher activity concentrations was assumed to extend throughout the swamp when calculating biota exposure, which the survey has indicated was highly conservative. The results of the survey can, therefore, provide a basis for establishing the spatial scale of impact. The productivity of plants in the area of highest contamination did not appear to be detrimentally affected [Yankovich et al., 2008b].

How the specific activity in sphagnum moss changed with distance from the area of highest C-14 specific activity was investigated and results indicated that there were essentially two groups of data. It was hypothesised that Group 1 was focused around areas of groundwater flow, whereas Group 2, which had lower activity concentrations, was less affected by groundwater flow, with activity concentrations being more associated with atmospheric transport. A reduction in C-14 activity in groundwater over time has been observed [Yankovich, 2008a; Killey et al., 1998] suggesting that volatilisation may have occurred, supporting the theory of atmospheric transport of C-14; overall, an order of magnitude decrease in the size of the most contaminated area has been observed between 1991 and 2001. However, an alternative explanation is that groundwater upwelling is occurring with sub-horizontal flow dilution, which could account for the observed reduction in C-14 over time.

Habitats have also been investigated at the site and their relationship with activity concentrations in groundwater investigated. Fen habitats, which can act as a barrier to groundwater flow over time, as organic matter accumulates, were associated with Group 1 results and their barrier characteristics could account for the retention of C-14 in such areas. Group 2 locations, on the other hand, were largely associated with marsh and swamp habitats, which can facilitate groundwater transport to the surface.

Paired samples of soil and vegetation were also analysed at some sampling locations and correlations between the specific activity in vegetation and in soils were observed. Samples of fauna were also taken and analysed to investigate how representative activity concentrations were in vegetation samples compared with the animals consuming them. Samples were collected from 6 sites, representing a range of possible C-14 exposure conditions and wetland habitats, as a means of investigating whether transfer to biota differed under varying exposure situations. It was hypothesised that animals would exhibit specific activities less than or equal to the foods that they consumed, with primary producers exhibiting the greatest specific activities. Overall, the results indicated that C-14 moss activity concentrations were quite representative of those in other biota. Amphibians were an exception, however, in that C-14 specific activities exceeded those of moss. This was particularly the case for frogs. This may be explained by amphibians inhabiting aquatic environments for a proportion of their time and/or obtaining some of their diet from the aquatic environment (or aquatic-derived prey, such as insects with aquatic life stages) for which activity concentrations are higher than in the terrestrial system.

Data derived from Duke Swamp have been used as the basis of a wetland inter-comparison exercise for the assessment of dose to non-human species within the IAEA’s EMRAS II programme.

3.3 SAMPLING EXPERIENCE IN A WETLAND ECOSYSTEM (DUKE SWAMP)

Liz Houser (Oregon State University) presented.

In the summer of 2012, a sampling programme was established at Duke Swamp. This focussed on aquatic areas and 10 water bodies were sampled in the period July to September. However, the sampling programme coincided with an interval during which no rainfall occurred over a 6 week period and a 100 year drought was declared. Overall, it appears there may be some trend toward dryer summers if climate change is real, which could have a large impact on wetlands and associated sampling programmes.

Duke Swamp itself is a high pH sphagnum peat bog that receives drainage from the waste management area, which has been active since 1963. The first detectable radioactivity in the Swamp was measured in the late 1960’s to early 1970’s. The source term has been poorly characterised although the major isotopes measured in groundwater are C-14 and H-3. It is estimated that the annual input of C-14 from the waste management area, in the form of CO_2, is some tens of GBq.

The Swamp is located in a low topographic region, below the waste management area. The area is characterised by the presence of spruce, cedar and maple trees and a high abundance of sphagnum moss as ground cover. Peat is present throughout the area at a depth of around 3 m. Due to its location in a low topographic area, minimal air mixing occurs except during storm periods.

There is a wide diversity of animals in the area, including beaver, a keystone species whose activities can greatly affect surface water characteristics – a potentially problematic issue when sampling in wetlands.

Whilst a large amount of work has been done to date in the region, little has been published and therefore remains unavailable unless specifically requested.

Further research work is however planned at Duke Swamp as input to a PhD project associated with Oregon State University and ideas are invited as to focus areas for this work.

3.4 SELLAFIELD DERIVED C-14 IN THE NORTH-EAST IRISH SEA

Pauline Gulliver (University of Glasgow), who was unable to attend the workshop, kindly provided a presentation on Sellafield derived C-14 in the north-east Irish Sea. The following section summarises the material provided.

The Sellafield reprocessing plant in north west England, began discharging to the Irish Sea in 1952. Initially, discharges of C-14 were relatively low (around 1-2 TBq/year) but have increased substantially since 1987 (Figure 3-5).

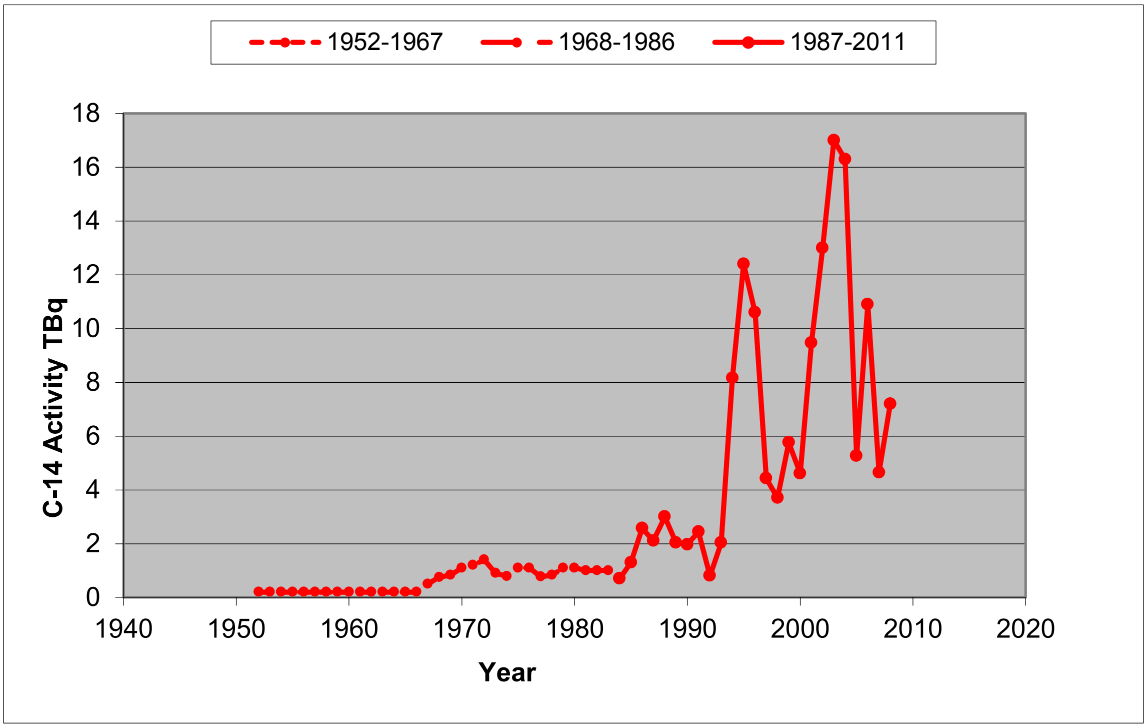


Figure 3-5. Discharge of C-14 from Sellafield to the Irish Sea (1952-2011).

Discharge of C-14 is in inorganic form and has an immediate impact on the C-14 activity of dissolved inorganic carbon (DIC) in NE Irish Sea. Following discharge, the C-14 behaves in a mainly conservative manner and is rapidly removed by prevailing currents through the North Channel although transfer to other carbon fractions of the water column (PIC, POC and DOC) is evident (Figure 3-6), as is uptake into biota. Activity concentrations of C-14 (net of ambient background 250 Bq kg^{-1} C) reported for the area are:

- Mussels: 1983±8 – 102±4 Bq kg^{-1} C;
- Seaweed: 592±3 - 44±3 Bq kg^{-1} C; and
- Shell material: 588±2 - 2161±6 Bq kg^{-1} C.

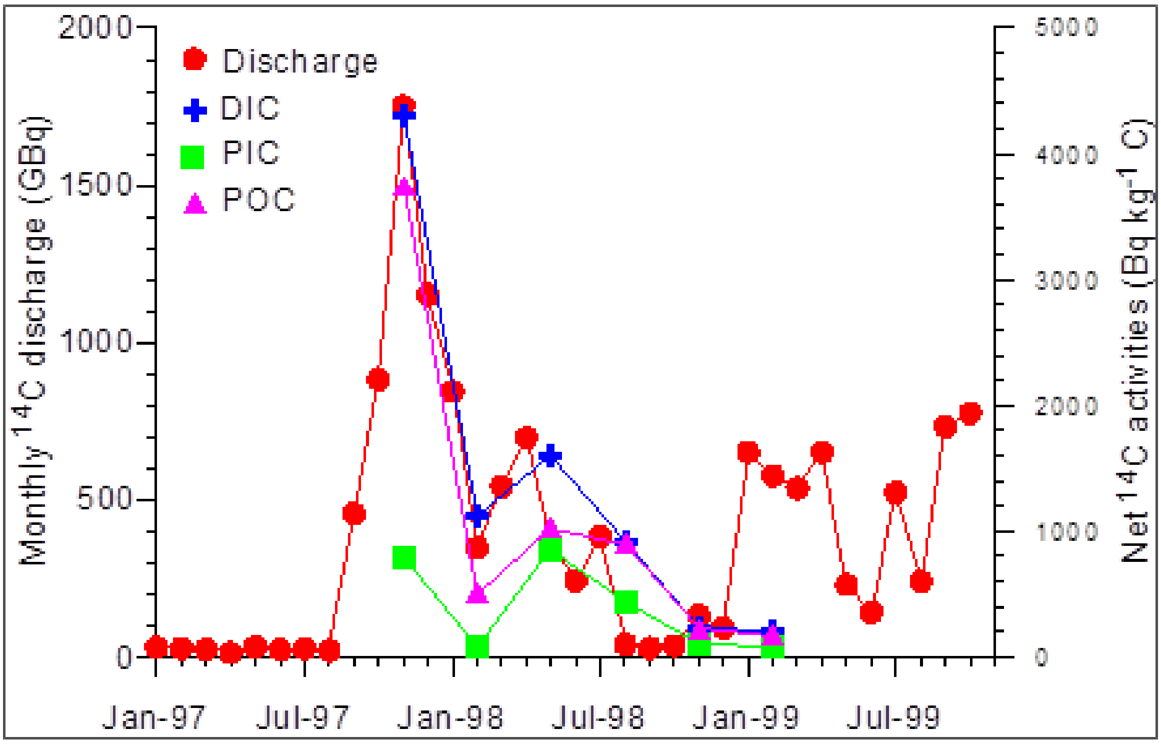


Figure 3-6. DIC, PIC and POC values for samples collected at St Bees Head in the NE Irish Sea, 1997-1999.

With death of biota, tissues eventually form the organic fraction of Irish Sea sediments while shell material is incorporated into the inorganic fraction of NE Irish Sea sediments: the organic sediment fraction shows clear incorporation of Sellafield C-14 in surface sediments with the inorganic sediment fraction showing lower activity (Figure 3-7). Inventories of approximately 917 GBq C-14 in the inorganic and 2045 GBq C-14 in the organic fraction of NE Irish Sea sediments have been estimated, which equates to only a small percentage of the discharged activity between 1952 and 1998. However, discharges have increased dramatically since the study was carried out in 1999. Prior to 1984, discharges were not reported. Nonetheless, total discharge estimates 1952-1998 range from approx. 76 -112 TBq whereas total C-14 activity discharged 1999-2011 was 113 TBq, suggesting that the budget of C-14 in Irish Sea sediments has doubled since 1999.

In addition to discharges from the Sellafield site, there is evidence of non-nuclear C-14 discharges occur on the east coast of the UK (Figure 3-8). These discharges result in high C-14 activities in biota compared with NE Irish Sea values, but in low DIC activities. One possible explanation for this discrepancy is that C-14 may be discharged in organic form.

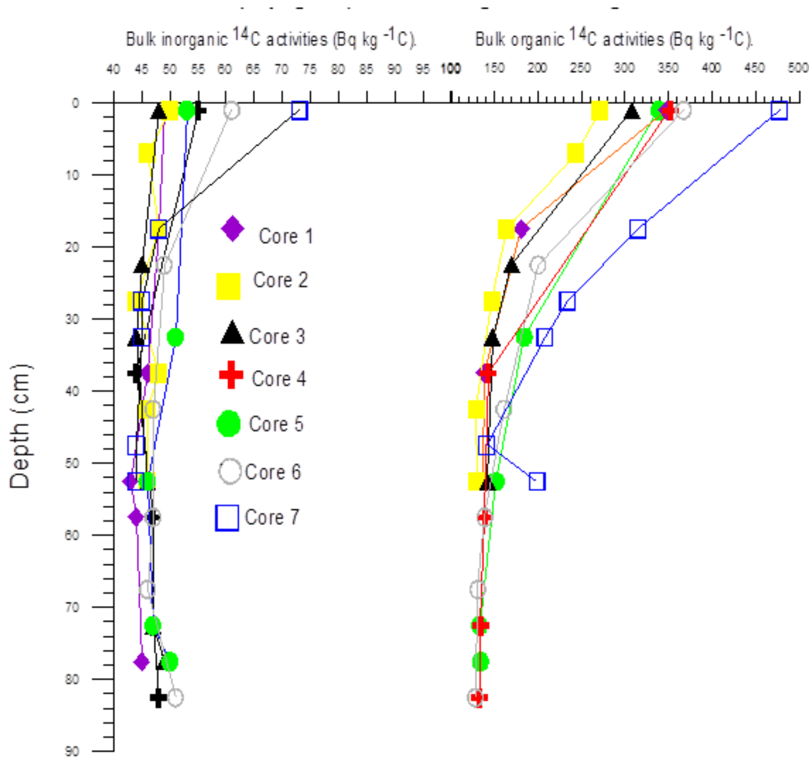


Figure 3-7. C-14 activity (Bq kg^{-1} C) in bulk inorganic and organic sediments.

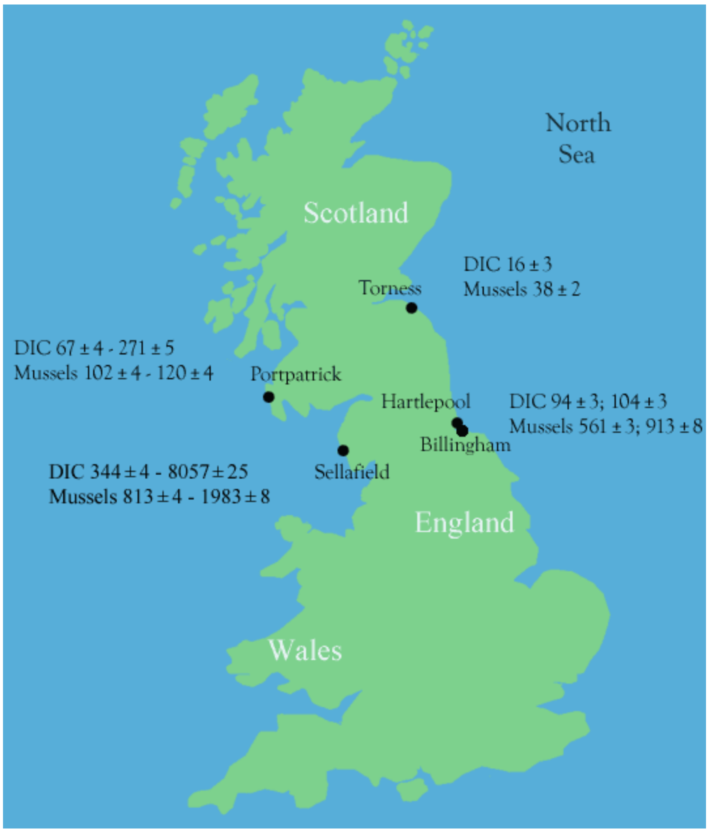


Figure 3-8. Comparison of C-14 activity concentrations in DIC and biota samples in the NE Irish Sea and off the east coast of the UK.

3.5 DISCUSSION POINTS

It was noted that data on C-14 behaviour in fish in rivers appears lacking, which is of interest to some organisations, notably NDA/RWMD. One discharge scenario considered in the generic approach adopted by NDA/RWMD to date has involved continuous discharge into a flowing system. In a slow water-exchange system, atmospheric loss is an effective mechanism for depleting the system of C-14. However, if the discharge occurs to a flowing water body then there are fast kinetics in the system that are dominant over atmospheric exchange. Since data on C-14 in rivers upon which model validation could be undertaken has been lacking, a conservative approach has been taken whereby high concentration ratios have been applied. Application of concentration ratios to calculate fish concentrations is not a realistic approach (concentration ratios require equilibrium conditions). Short-term dynamics between inorganic and organic carbon need to be more readily understood therefore. Questions around C-14 behaviour in flowing systems are therefore different from those for lake or wetland systems, where turnover between water and sediments is more significant than overall water loss. Considerations for flowing systems need to consider rivers of a sufficiently large size and of moderate enough flow to sustain fish populations.

UK data on P-32/P-33 in rivers, inclusive of kinetic data, could be used as the basis of a comparison. Data on C-14 may be available from the Rhone River in France and it is a well characterised river which also receives discharges from a number of nuclear sites. If data on carbon and C-14 dynamics in the Rhone, or alternative large river systems are available, these could provide the basis of a model evaluation exercise.

Whilst river data may be lacking, marine data are available for C-14 in fish, although fish samples have largely been bulked for analysis such that information on fish age groups is not available. Work is however on-going by EdF to analyse fish individually and these data may be available in the next 12 months or so.

In relation to Duke Swamp, it was noted that there appear to be sufficient data to enable a hydrological model of the swamp to be developed, which would allow atmospheric losses of C-14 (for example net respiration losses) to be investigated, by comparing the observed concentration distribution with model simulations with different loss rates from the superficial layer of active vegetation. From the data presented it appears that a 3-D plume has developed from the point source release that is diluted as it expands. It was noted that MIKE-SHE could be a suitable platform for model development. However, it is uncertain as to whether such model development has already been progressed, using this or alternative tools, by hydrologists at AECL.

It was also noted in relation to the presentations from Duke Swamp that the data could be of interest to the BIOPROTA geosphere-biosphere interface (GBI) project since it is evident that discharge occurs to a small area with subsequent distribution in a much greater area. Mixing has not been immediate. The data may therefore, with further analysis, be useful as input to understanding long-term behaviour of C-14 in wetlands and informing assessment model development.

4. WHOLE SYSTEM APPROACH TO C-14 MODELLING

Following on from the presentations on the plant-canopy-atmosphere in the terrestrial system and the C-14 modelling in the aquatic environment, the whole system approach to C-14 modelling was addressed. As an introduction, the results of a study to identify and quantify pools and fluxes of both living and dead organic matter in a landscape in Sweden, Kumblad et al [2006], were summarised.

The study found that the water exchange between coastal basins and between the basins and the open Baltic is the single most important factor determining the carbon dynamics of the coastal basins, at least in the outer basins where carbon transported by water currents contributed more than 90% of the total carbon input. Terrestrial systems have the greatest pools of carbon (soil organic carbon – SOC) and, in larger catchment areas, terrestrial sources can account for around 20 % of total carbon input to the local marine environment. Results indicate that it is the biotic processes (primary production and respiration) that strongly dominate the flux of matter in terrestrial environments, whereas abiotic processes (import and export of matter due to water exchange) are completely dominant in the outer marine basins. Modelling results also indicate that it is the inner marine basins and lakes that have the highest potential to accumulate radionuclides to high levels. These basins have much slower water turnover than the rest of the marine basins, they show intermediate to high rates of detrital accumulation (receiving a comparatively large amount of carbon as discharge from terrestrial catchments), and they are also likely sites for discharge of radionuclides in the case of leakage from a deep repository. The study indicates, at least for a boreal landscape mosaic of the type investigated, that the highest potential for accumulation of discharged radionuclides can be found on accumulation bottoms in aquatic environments with slow to moderate water turnover. These shallow near-land marine basins may constitute a potential risk for exposure to humans in a future landscape as, due to post-glacial land uplift, previous accumulation bottoms are likely to be used for future agricultural purposes.

4.1 C-14 WASTE FORM AND EFFECTS ON RELEASE

Klas Källstrom (SKB) presented.

The physics governing the production of C-14 in nuclear power reactors is mainly associated with three nuclear reactions: $^{14}N(n, p)^{14}C$, $^{13}C(n, \gamma)^{14}C$, and $^{17}O(n,\alpha)^{14}C$. The reaction with ^{17}O in H_2O produces 23-24 kBq/MWh(Th) in a BWR, and 15-16 kBq/MWh(Th) in a PWR. It will be produced in the form of CO_2 and CH_4 and other species. More than 94% will be discharged to atmosphere, and the rest (0.5% to 6%) will be incorporated in ion-exchange resins, placed in waste packages and sent to SFR.

The chemical form of C-14 in the ion exchange resins has been investigated, see Table 4.1. The majority is inorganic, in the form of bicarbonate.

Table 4.1. Chemical form of C-14 in ion-exchange resins (F indicates Forsmark, O indicates Oskarsham, B indicates Barsebäck and R indicates Ringhals).

	Accumulation, % of production		
Unit(s)	Total	Inorganic	Organic
F1+F2	1.1	1	0.1
F3	2	1.9	0.1
O1+O2	0.39	0.38	0.01
O3	0.91	0.9	0.01
B1+B2	0.39	0.38	0.01
R1	4.7	4.6	0.1
R2+R3+R4	5.6	4.1	1.5

The ion exchange resins are placed in a filter flush tank, stored and then immobilised. The initial C-14 content in the store at the reactor is 5-8%. The liquid is then transported to a store near the encapsulation plant. The C-14 content of liquid from this store, which goes to the encapsulation plant, is about 1%. The exact value depends on how it is stored and for how long. It is therefore important to obtain information on the C-14 content as close to the waste immobilisation plant as possible. The activity and mass of C-14 in SFR is given in Table 4.2.

Table 4.2. Activity and mass of C-14 in the SFR by disposal location (see Section 4.2)

	Activity in SFR at closure (Bq)			mass of C-14 in SFR at closure (g)		
Unit(s)	Total	Inorganic	Organic	Total	Inorganic	Organic
Silo	3.0E+12	2.4E+12	6.3E+11	18	14	4
1BMA	1.5E+12	1.4E+12	1.4E+11	9	8	1
XBMA	3.1E+11	3.0E+11	1.6E+10	2	2	0.1
1BTF	2.4E+11	2.3E+11	1.1E+10	1	1	0.1
2BTF	2.2E+11	2.1E+11	5.3E+09	1	1	0.03
1BLA	4.2E+09	4.1E+09	8.2E+07	0,03	0,03	0.0005
XBLA	5.2E+09	1.7E+09	3.5E+09	0,03	0,01	0.02
BRT	9.8E+09	-	9.8E+09	0,07	-	0.07

Hence the total mass of C-14 disposed in the SFR is about 30 g. This is based on measurements of resins for 4 or 5 fuel cycles. The organic form will not sorb onto the barriers, whereas the inorganic form will be sorbed (through reactions with carbonate). It is unlikely that all inorganic C-14 will be released as methane.

4.2 C-14 DOSE ASSESSMENT FOR SFR

Eva Andersson (SKB) presented.

SFR, an existing repository for low and intermediate waste, was constructed in the 1980s. It contains a silo and a series of tunnels (1BTF, 2BTF, BLA and BMA). The total storage capacity is 63,000 m^3 (~400,000 m^3 excavated volume) and the total allowed activity is 10^{16} Bq. The silo is intended to host 80% of the activity in SFR (and ~20% will be in BMA).

The results from the previous safety assessment SAR 08 indicated that the dose from a lake system at about 5000 years after present was dominated by C-14. An extension to SFR is planned and hence biosphere work on safety assessment for the planned extension of SFR has to give emphasis to C-14.

The main factors affecting the dose are landscape evolution and the modelling of movement of radionuclides in the ecosystem. Potential release from the repository will occur to low-lying areas in the landscape. The landscape has been divided into a series of biosphere objects where release from the repository can affect humans and the environment, representing sea basins, bays, lakes and mires. The landscape of Forsmark will change in the future due to land-rise and succession and marine basins will be transformed to lakes and mires which may later be used as agricultural land. The new assessment has updated the model with new site data and considered the utilisation of the landscape. Three self-sustainable communities have been considered: hay from contaminated mire for infield-outland farmers based on Iron Age (500 BC – AD 1000) farms; drained wetland/lake - Industrial age agriculture (~1900); and hunter and gatherers foraging the most contaminated land based on stone-age settlements in Central Eastern Sweden found near the coast line. Traditional diets have been considered for these communities.

The model was reviewed and conservatisms and possibilities for improvement were identified as the atmospheric model, the transition between ecosystems and the chemical form and behaviour of the carbon input (e.g. methane). In the previous assessment, all the methane was assumed to be converted to CO_2. However, some may be released directly to the atmosphere as gas bubbles through methane ebullition.

CO_2 uptake occurs in the summer and the previous model considered annual means. Preliminary modelling with higher resolution in time has been performed. Concentrations in primary producers and in surface waters show a similar seasonal pattern of fluctuations, whereas the fluxes in pore water in till show the opposite pattern. However, evaluation of the effect of higher resolution on the end result has still to be made. Dilution of aquatic C with terrestrial C is another factor that has been considered. Uptake in fish is calculated from the C-14 concentration in water assuming that the food web is based on primary production within the contaminated lake. However, many lake food webs are dependent on terrestrial carbon and not in-lake produced carbon. Hence, if C-14 discharges to the lakes, there could be a dilution in the food web due to the input of ’uncontaminated’ terrestrial carbon. Snow melt and rainfall can also contribute to high C inflow, and consequent dilution.

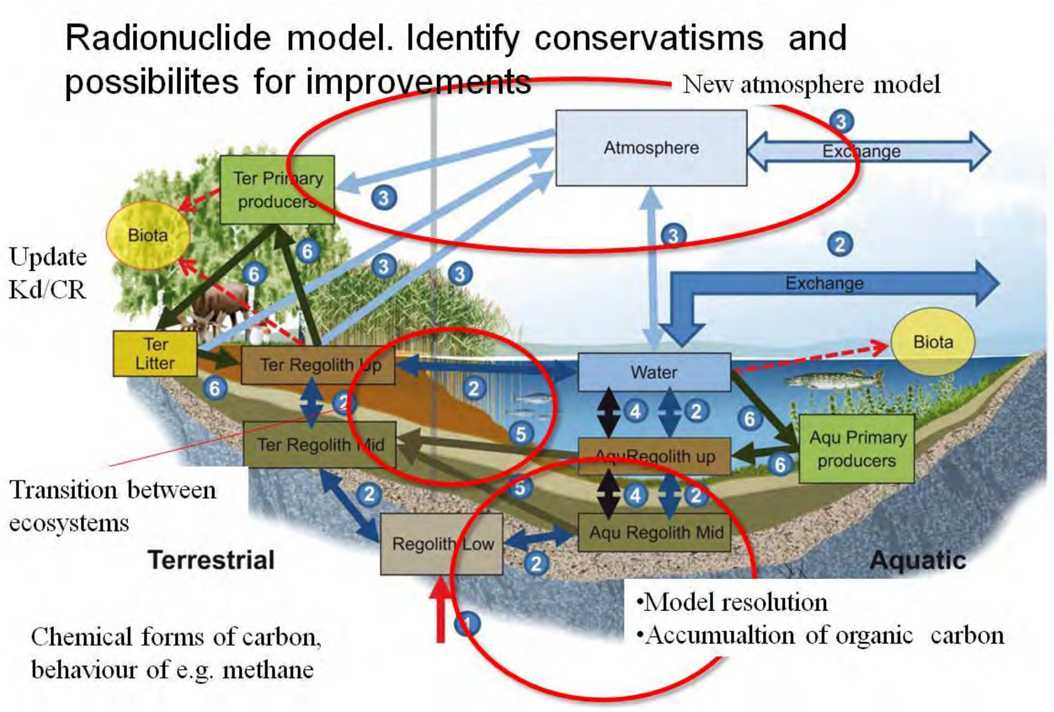


Figure 4-1 Chemical forms of carbon - Transformation between organic and organic carbon, modified figure from SKB [2010].

The main possible model modifications being considered as relevant to C-14 are related to:

- Human land use and diets;
- The accumulation of C in peat and sediments;
- The chemical form and oxidation of CH_4 to CO_2;
- The effects of seasonal variations on C-14 transfers in the lake food web;
- The effect of dilution with terrestrial carbon on the C-14 in the lake food web.

4.3 C-14 MODEL DEVELOPMENT AT SKB

Ulrik Kautsky (SKB) presented.

C-14 measurement methods have improved over the years, resulting in skills such as understanding of compensation factors (to correct for the variation in $^{14}C/^{12}C$ ratio) being forgotten and hence the old data cannot be interpreted with ease. Carbon models are standard in ecology and provide all the information needed on movement through the system. The flux of C-14 through water movement in the study area Oregrundsgrepen is very large. Once the carbon pools are understood, then it is possible to map C-14 onto the C pool and flux model. An extensive measurement programme has been undertaken.

The food pathway is the important route for modelling the impact of C-14 disposed of at SFR. The transfer factor (TF) into the food (for example. fish) is therefore important. Measurements of TF at the study site differ from IAEA values (mainly laboratory measurements) since they incorporate the impact of the C-14 flux that is transported through water movement. The coastal fish community is dominated by herring and sprat (60-70 kg/ha), whereas in the inner bays, perch, roach and white bream dominate. Variations in shore level, permafrost, and ice cover are considered over several sequences. Different sites are used as analogues for the different climate states e.g. a site in Greenland for the permafrost analogue.

Hydrodynamical modelling of past climate states has been performed using the Mike 3 model available from the Danish Hydraulics Institute (DHI) [Karlsson, et al. 2010].

A complex box model has been developed to explore processes. This then informs transfers in the assessment model. The radionuclide model is shown in Figure 4-2.

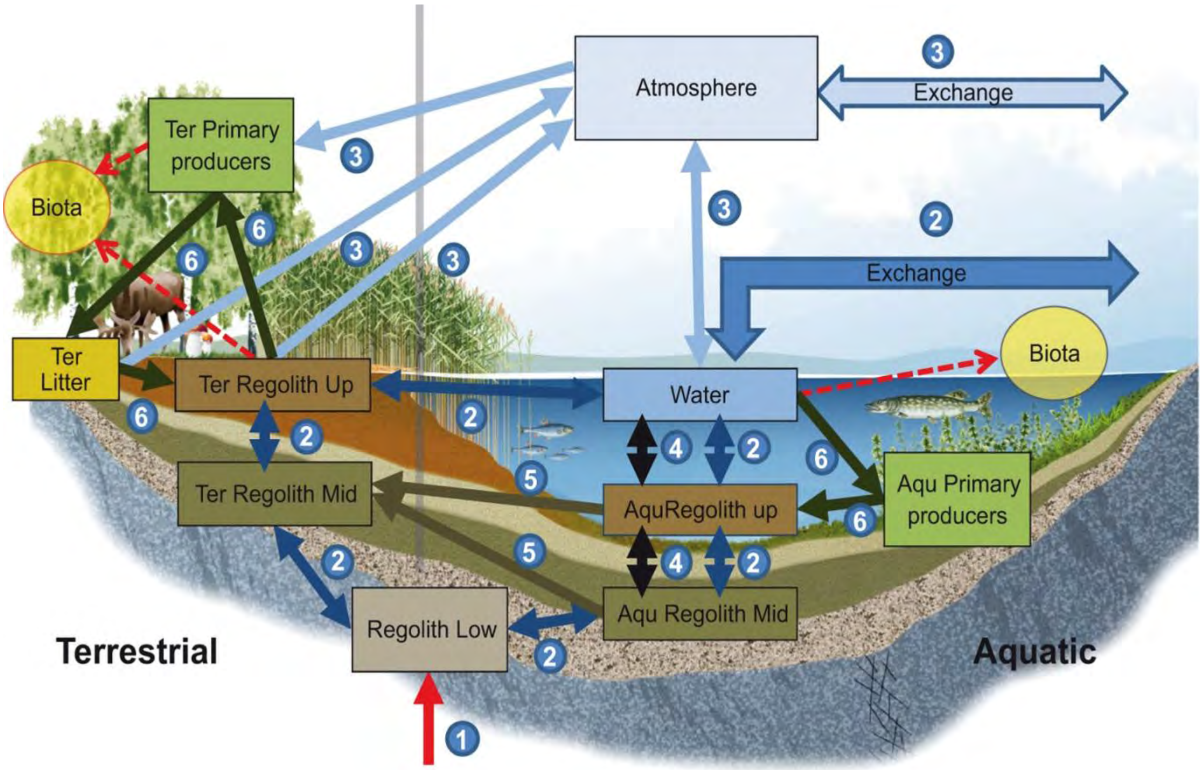


Figure 4-2. Radionuclide model for C-14 transfer in terrestrial and aquatic environments, Figure from SKB [2010]

The model can represent changing transfers with time, accumulation in peat and transfer of material to the terrestrial compartment.

Carbon and nutrient turnover are well understood processes. For other elements, related models can be constructed based on discrimination factors obtained from site-specific data. Such models can be calibrated and validated for present conditions, and then projected for future conditions. Confidence is built by looking at uncertainties and sensitivities, and by checking mass balances.

4.4 CARBON-14 BACKGROUND, PATHWAY, AND DOSE OPTIMISATION ANALYSIS

Emily Caffrey (OSU) presented.

The first phase of the project was to compile an inventory of C-14 (at US NPP) and this has been completed. The second phase is to review human exposure pathways. The majority of sites use a standard NRC model and parameter values for the human dose assessment because it is easy.

However, some of the parameters require updating. The current model RG 1.109 (1977) is based on ICRP Publication 2 for human dosimetry and is, therefore, completely outdated.

The first parameter to be reviewed was the plant uptake from the atmosphere. RG 1.109 equation C-8 defines the fractional equilibrium ratio. This is defined as the ratio of total annual release time to total annual time during which photosynthesis occurs. Continuous releases are assigned a value of unity. However, the equation is based on an annual photosynthesis time of 4400 hours (~all daylight hours) whereas actual photosynthesis times will be less: for corn ~1440 hours, soybeans ~1080 hours and lettuce ~700-800 hours.

The assumptions for the release rate (Q14) in RG 1.109 are continuous release, steady state meteorology, environmental transport compartments are at equilibrium and uptake is continuous. In reality, physical transport of C-14 is transient, steady concentrations of CO_2 do not exist in environmental reservoirs, only C-14 released during the daylight growing season will be taken up by vegetation, CO_2 fixation in plants occurs quickly, uptake rates varies diurnally and seasonally, and the incorporation rate into vegetation tracks short-term atmospheric concentrations.

The plant carbon fraction in RG 1.109 is 0.11, based on the assumption that C-14 is only incorporated into vegetation as CO_2 and ~100% of carbon in vegetation comes from the atmosphere as a result of photosynthesis. However, the rate of uptake in vegetation varies considerably with meteorology since drought and other environmental stressors can shut down CO_2 intake and utilisation. Photorespiration was not considered in RG 1.109; this will be considered further.

Carbon concentration in edible plant portions changes during the plant growth cycle, and some C-14 can be transported to inedible portions of the plant. A biological half-life of carbon in plants of 50 days is proposed by Collins and Bell [2001].

The natural atmospheric carbon concentration in Equation C-8 in RG 1.109 is assumed to be 0.16 g m^{-3}, whereas today's value is closer to 0.19 g m^{-3}, due to the Suess Effect.

The food chain transfer parameters (days/L milk ingested by the animal) used in RG 1.109 were compared with Galeru et al. 2007, see Table 4.3.

Table 4.3 Food chain transfer parameters used in RG 1.109 compared with those from Galeriu et al. 2007.

Product	USNRC Transfer Coefficient	Galeriu et al. 2007 derived transfer coefficient
Milk (d/L)		
Cow	0.012	0.011
Goat	0.10	0.067
Meat (d/kg)	0.031 (type not specified)	0.046 (beef)

The next steps are to review the inhalation rates [ICRP 23 vs. 103], food-stuff consumption rates, and vegetation uptake factors. IAEA Report 472 [2010] Tables 5-7 and 5-8 have vegetation-dependent uptake factors that vary considerably from the generic value used in RG 1.109. A review of C-14 plant uptake models (e.g. the models discussed in BIOPROTA) will also be carried out.

The third phase, also completed, was to review the effectiveness of vegetation analysis using liquid scintillation counting (and compared to Accelerator Mass Spectrometry - AMS). AMS is an exceptionally sensitive technique for measuring concentrations of isotopes in small samples, typically less than 1 milligram, and the relative abundance of isotopes at low levels. The resulting sensitivity is typically a million times greater than that of conventional isotopic detection. For biological studies, AMS has been used primarily for counting carbon-14 because carbon is present in most molecules of biological interest and carbon-14 is relatively rare in the biosphere. Tritium (hydrogen-3) has also been used extensively as a tracer in biological research. The use of tritium in AMS is new and holds great promise, because many molecules are easier to tag with tritium than with carbon-14. Other isotopes that can be measured by AMS include plutonium-239, calcium-41, beryllium-10, chlorine-36, and iodine-129. Samples of green vegetables were collected from a control garden and two downwind gardens. No difference was found between the control garden and sample gardens (and the same result was reported by the AMS laboratory).

The presenter referred to the following references: Aquilonius, K. & Hallberg, B. [2005], Aulagnier, et al. [2012], Aulagnier et al. [2013], Dias et al. [2008], Le Dizès [2005], Le Dizès, et al. [2012], Galeriu et al. [2007], Limer et al. [2011], Roussel-Debet et al. [2006], Takahashi et al. [2011], USNRC [1977], Xu et al. [2011], Yim and Caron [2006], Collins and Bell [2001], Fischer et al. [2007]. Key, Killough and Rohwer [1978].

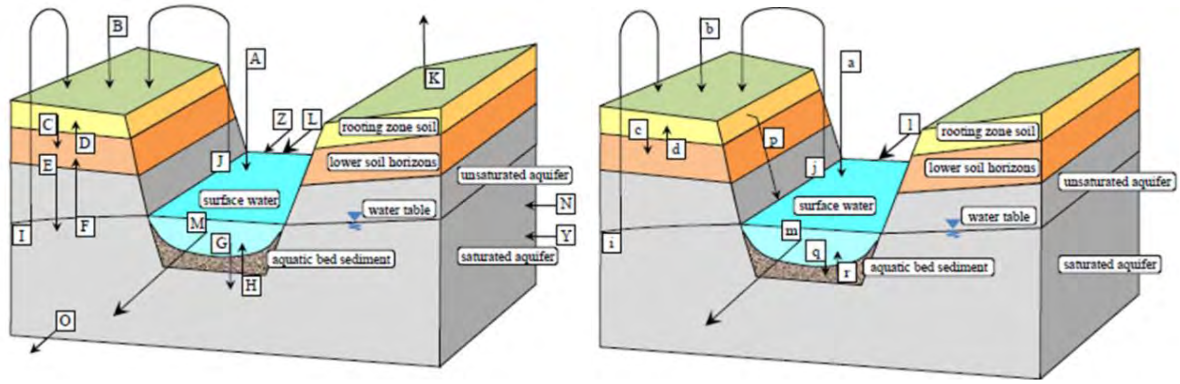
4.5 NAGRA C-14 DOSE ASSESSMENT

Sven Keesman (Nagra) presented.

Stage 2 of the Swiss site selection plan for deep geological repositories for L/ILW and for HLW requires a so-called provisional safety analyses. Two new codes have been developed for the assessment of the biosphere. The main NAGRA biosphere code, SwiBAC [Walke and Keesman, 2013], is a re-implementation and extension of the former code TAME [Klos et al, 1996]. The other is an implementation of Nagra's model for the assessment of C-14 in the biosphere [Nagra, 2013].

The conceptual model of the biosphere is an inland biosphere situated on a river valley. A range of river types have to be considered from small streams to large regional rivers for the assessment of the six siting regions.

The release from near-field and transport through the geosphere to the biosphere, i.e. to a shallow aquifer or surface water, takes place on a long time scale. Exposure of humans due to the release happens by ingestion of agricultural products, drinking water, inhalation of dust and direct exposure from the ground. Food supply is provided by a fully self-sustaining agricultural system producing crops and fruit, meat, eggs and dairy products and which does not cause losses of radionuclides from the system. Radionuclides disappear from the system only by radioactive decay or by outflow of ground or surface water. Radionuclide transport between soils and water bodies is modelled using a dynamic approach in a compartmental structure. The conceptualisation of relevant fluxes of water and solid material within a section of the biosphere is shown in Figure 4-3 and the (simplified) derived compartmental structure in Figure 4-4.



a) b)

Figure 4-3. Conceptual fluxes of water (a) and of solid material (b) represented in SwiBAC

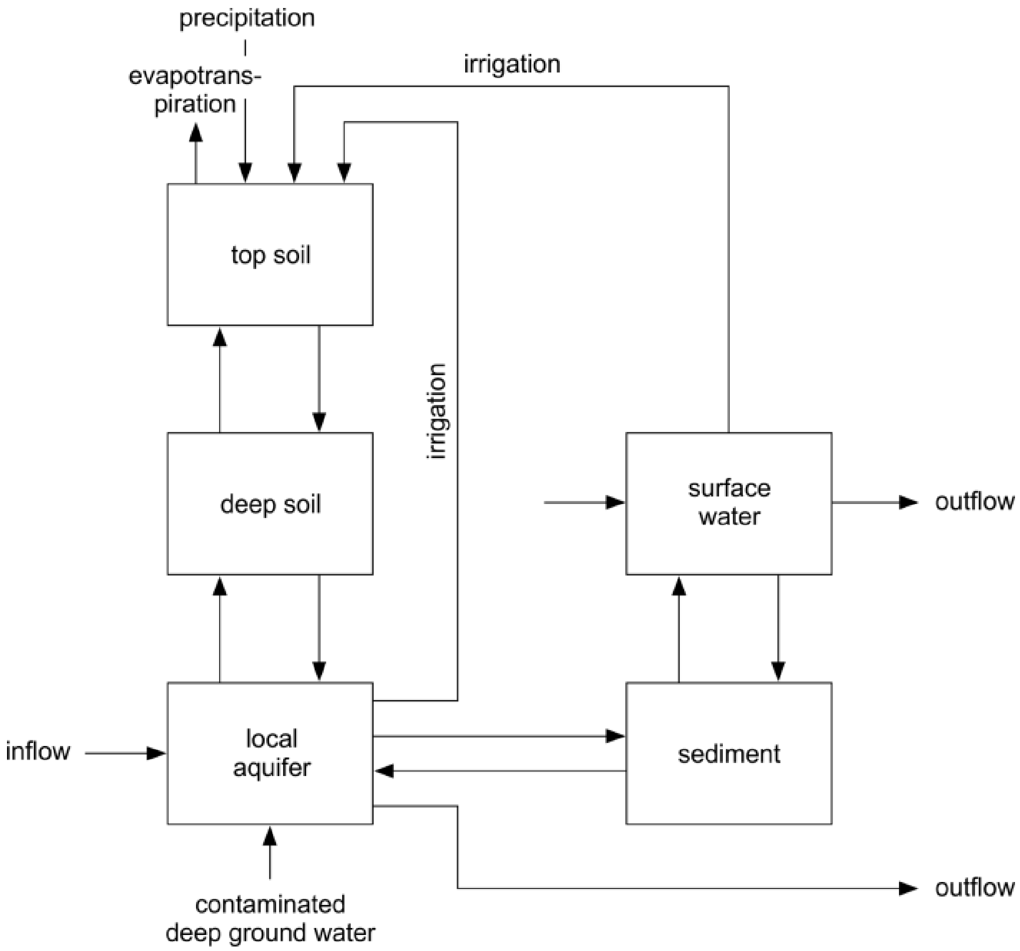


Figure 4-4. Compartmental structure of the dynamic transport model in SwiBAC

Transfer of radionuclides from soil and water to crops, livestock and humans is modelled using an equilibrium approach with five different crop types being considered: green vegetables, root vegetables, cereals, pasture and fruit (Figure 4-5).

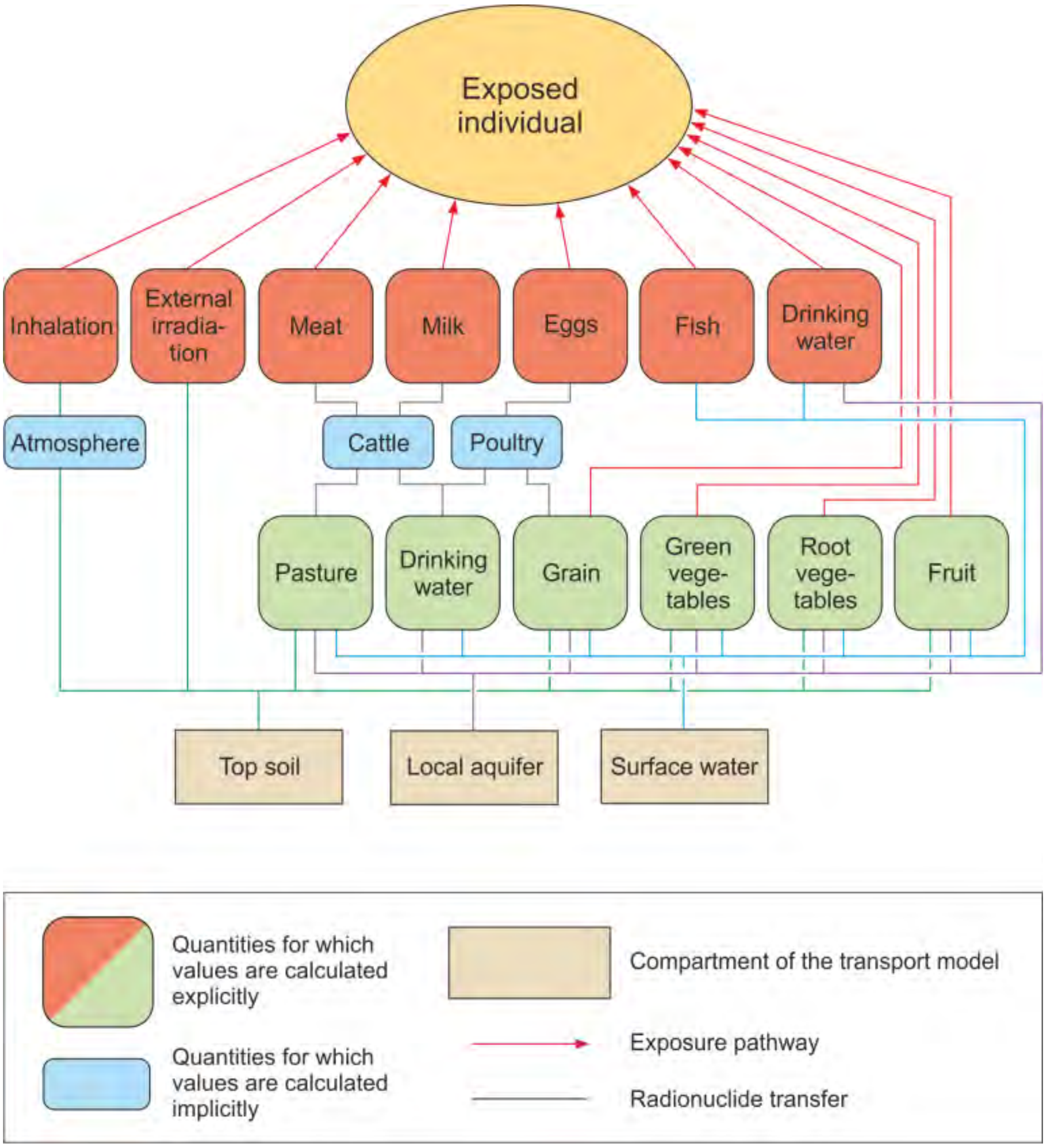


Figure 4-5. Pathways of exposure considered in SwiBAC.

The SwiBAC model is run for a constant release rate to steady state to determine a biosphere conversion factor for each dose relevant radionuclide. Different climate states have to be considered: present-day reference case, warmer and drier, and periglacial (see [Nagra 2010]).

The Nagra C-14 Model (NC14M) [Nagra, 2013] describes the transport of C-14 in the biosphere based on a specific activity approach, i.e. relative to the amount and fluxes of stable carbon in and between the considered pools of carbon. It has been developed in order to derive parameter values for SwiBAC for C-14 for the dose assessment. This approach aims to consider the special aspects of C-14 in the biosphere and maintain a uniform model for the dose assessment of all radionuclides at the same time for the provisional safety analysis of the different siting regions. It is based on the C-14 model used in Stage 1 [Brennwald and van Dorp 2008] describing a terrestrial plant-soil-atmosphere system. The C-14 released from the near field of the repository is assumed to be either dissolved in water (CO_2, CH_4, organic) or to be released as gas (CO_2, CH_4). All gas is assumed to enter the deep aquifer where it dissolves in the ground water. It is assumed that all CH_4 is oxidised to CO_2 and, as such, available for uptake by plants. The processes included in the model are: exchange of carbon in soil (organic, inorganic, gas); exchange of gas in soil with atmosphere (atmospheric pumping); atmospheric canopy dynamics; photosynthesis (assimilation of atmospheric CO_2 by plant); respiration (plant to atmosphere/soil); degradation/decomposition of plant material (in soil); uptake and exudation of inorganic carbon by plant from/to soil; and harvest of plants. The distribution of stable carbon is in steady state, i.e. there is no accumulation or depletion of stable carbon in the different carbon pools. The compartmental structure of NC14M model and the considered exchange processes are shown in Figure 4-6.

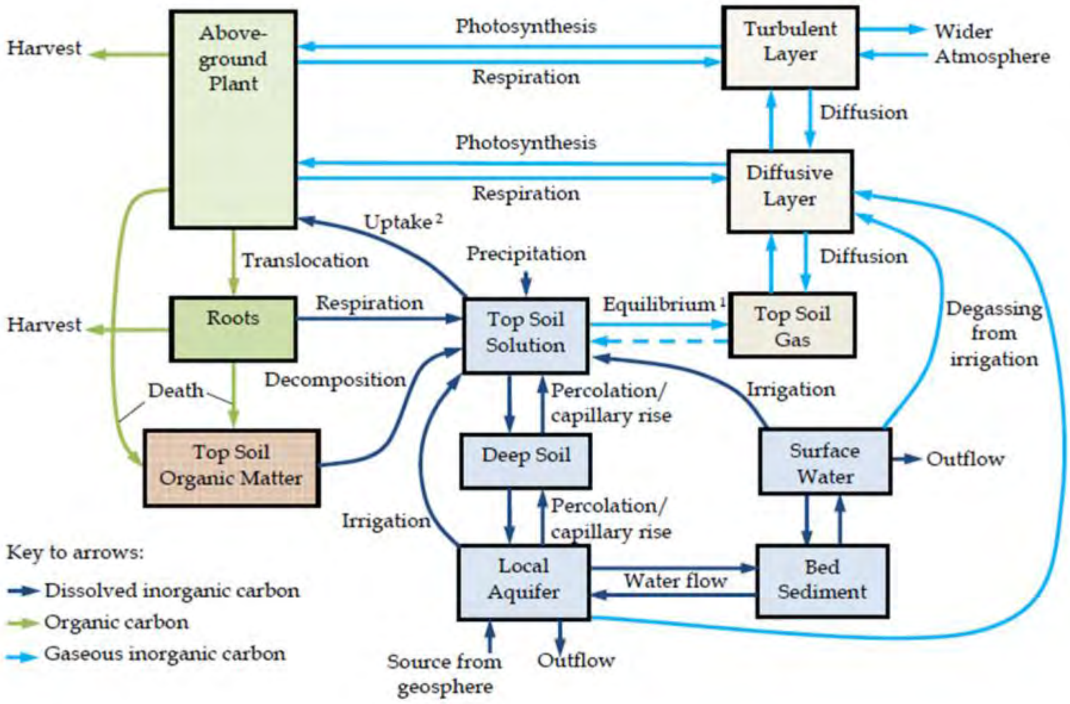


Figure 4-6. Compartmental structure of NC14M

Apart from 'Top Soil' all SwiBAC compartments correspond to a single carbon pool (and accordingly a single compartment) in NC14M; 'Top Soil' splits into three different carbon pools for organic matter, soil solution and gas. For the representation of the plant and the atmosphere two additional compartments are introduced in NC14M, as depicted in Figure 4-7 ('Above Ground Plant' / 'Roots' and 'Diffusive Layer' / 'Turbulent Layer' respectively). In contrast to SwiBAC only one single crop type is being considered at a time. The transfers of C-14 between compartments (λ_{C14}) are derived from fluxes and amounts of stable carbon by:

λ_{C14} = flux of stable carbon / amount of stable carbon (in donor compartment).

The size of compartments, crop yields and harvesting rates in NC14M are derived from SwiBAC, and the carbon fluxes in NC14M are derived from water fluxes in SwiBAC (as applicable). Integration of NC14M into SwiBAC is achieved by using derived C-14-parameters in SwiBAC. Losses of C-14 from the NC14M-system (harvest, atmospheric losses) can be represented by an extra loss term in SwiBAC obtained by balancing the top soil compartments with the plant and atmosphere compartments of NC14M. An effective sorption coefficient for soil (K_d) can be derived from the identification of transfer rates in SwiBAC with rates in NC14M:

$$\lambda_{NC14M} = \lambda_{\mathrm{SwiBAC}} = \frac{\text{water flux}}{(\text{porosity} + \text{density} \times K_d)}$$

The value of K_d for C-14 for the top soil is dependent on the crop type. By means of these parameters, the distribution of C-14 derived in NC14M can be reproduced in SwiBAC.

An effective soil-to-plant concentration ratio (CR) is used to determine the uptake of C-14 by the crop from soil, where:

$$CR = \frac{C_{Plant,AboveGround}}{C_{TopSoil}} \quad \text{or} \quad CR = \frac{C_{Plant,Root}}{C_{TopSoil}}$$

depending on crop type. A value for SwiBAC can be specified by defining a generic crop type or by taking an average of all crop types.

Climate specific parameters such as precipitation, irrigation, groundwater flow and surface water flow have a limited effect on the steady state dynamics of loss from the soil to atmosphere and uptake by plants and such on the CR, K_d, and loss rate from the soil. However, crop-dependent parameters (yield, canopy height, water content) do affect uptake dynamics and the effective CR, changing the loss rate from Top Soil.

Conservatively, no advection is currently assumed in the lower canopy layer (the diffusive layer) in NC14M, though this is under review. Increased atmospheric advection will reduce the value of CR and the corresponding biosphere conversion factors.

4.6 LONG TERM ASSESSMENTS, SHORT TERM DATA: CAN ONE MODEL BE APPLIED TO BOTH?

Ryk Klos (Aleksandria Sciences, for SSM) presented.

SSM has a regulatory requirement to develop in-house modelling of C-14 for oversight purposes. The model needs to address chronic releases in groundwater, both chronic and acute gaseous releases to the soil, and both chronic and acute releases to atmosphere, and include soils, plants and atmosphere. The 2011 C-14 project included a qualitative review of models and modelling approaches, followed by scoping options for model development.

The model review included:

- Operational and accidental release models applicable to the atmosphere & surface water, routine releases and accidental releases: STAR, PRISM, POM C-14, N288.1, OURSON, and TOCATTA , and models developed by both KAERI and NIRS were considered, and;
- Waste disposal models (deep and shallow radioactive waste disposal): enhanced RIMERS model, Avila and Pröhl (developed for SKB and applied at Forsmark), AquaC_14 and Thorne-Limer model used for the LLWR 2011 Environmental Safety Case (ESC).

The general observations relating to the concepts used in these two different types of model are summarised in Table 4.4.

The plant review considered the following:

- Photosynthesis, particularly the canopy – atmosphere interaction (diffusion in the lower canopy, turbulence within the upper canopy and turbulent ‘free air’);
- Leaf area index;
- Windspeed;

to determine ‘what “air” the leaves see?’ The review also considered root uptake (factors of 1% to 2% are generally accepted, but it could be up to 5%).

Table 4.4 Review of the FEPs included in operational and waste disposal models.

FEP	Operational Models	Waste Disposal Models
Time steps	• Often sub-annual	• Equilibrium conditions or annual average
Source	• Gas from aboveground • Irrigation water	• Gas from belowground • Upwelling water • Irrigation
Soil	• Not always explicitly modelled • One or multiple compartments	• Not always explicitly modelled • One or multiple compartments
Plant	• Often multiple compartments • Dynamic plant growth • Isotopic dilution (growth)	• Typically a single compartment • Static plant biomass • No isotopic dilution
Atmosphere	• Sometimes multiple compartments	• Sometimes multiple compartments
Plant ^{14}C concentration	• Specific activity approach (photosynthesis)	• Specific activity approach (photosynthesis) • Root uptake?

The review of contemporary models and a parallel conceptual model review were synthesised into a new model called SSPAM^{14}C (Swedish Soil-Plant-Atmosphere Model for C-14) implemented in ECOLEGO. The model contains 6 soil compartments, 3 plant compartments, 2 atmospheric compartments and a sink, see Figure 4-6.

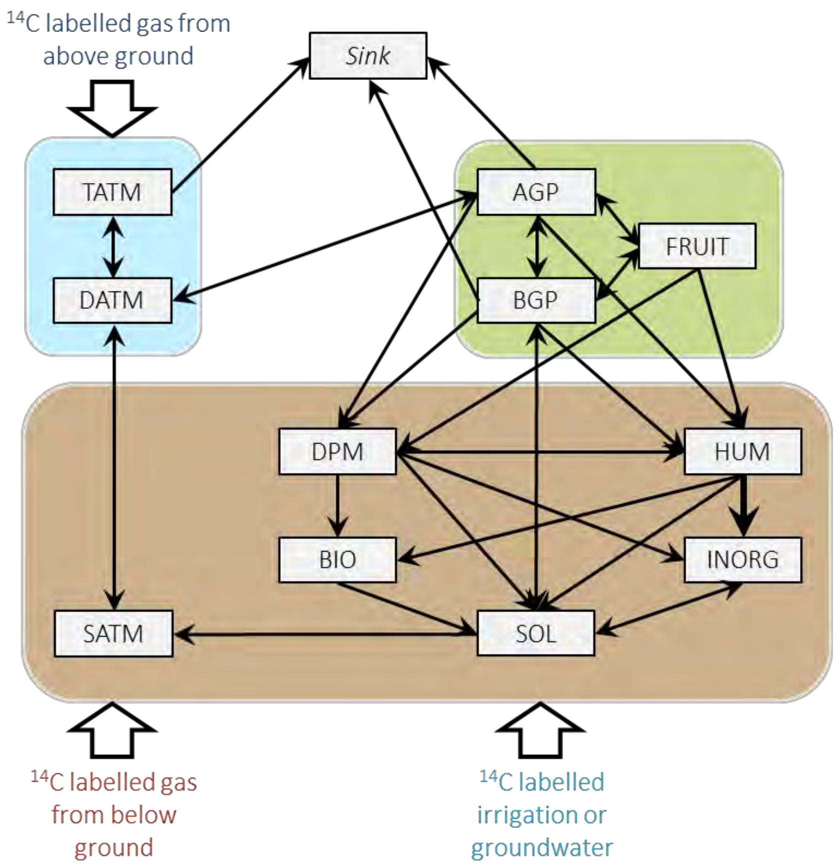


Figure 4-6. SSPAM^{14}C (Swedish Soil-Plant-Atmosphere Model for C-14).

The transfers within the model are defined by observable parameters and the source term can be to soil solution (SOL), soil atmosphere (SATM), or above-ground atmosphere (TATM). The model includes (inter)annual processes, static or dynamic plant biomass and a root uptake option. The model was tested against an implementation of Enhanced RIMERS derived from the published model, for a release to SOL (soil solution). Reasonable agreement for crop content was obtained, see Figure 4-7.

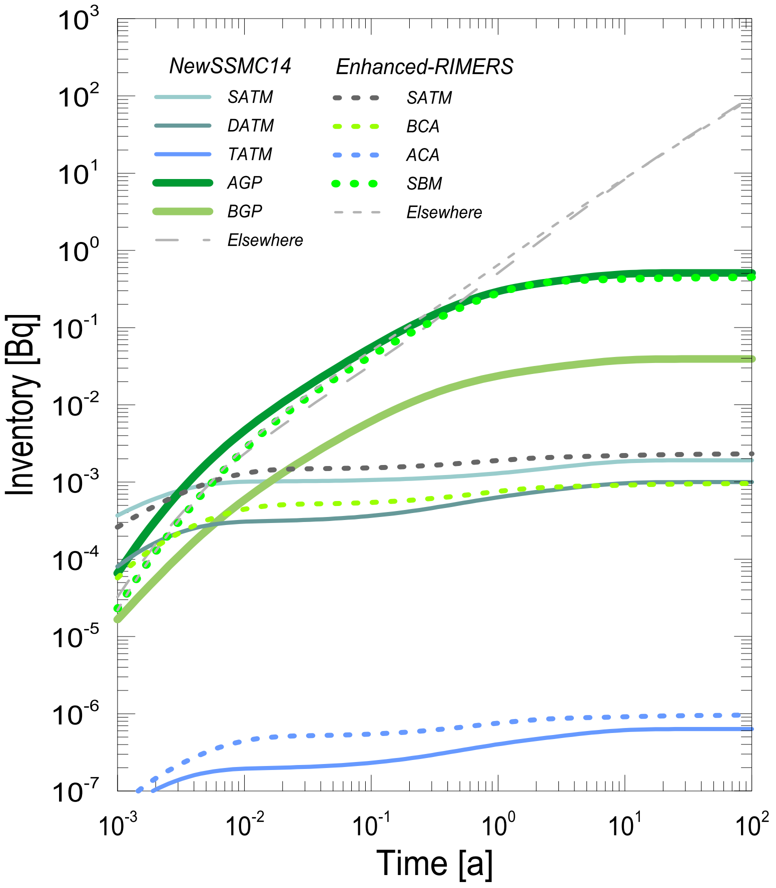


Figure 4-7. SSPAM^{14}C and Enhanced-RIMERS model comparisons for release to a soil solution

Validation was performed by considering the experiments carried out at Imperial College in the 1990s [Tucker and Shaw, 1997]. In a study for the UK Ministry of Agriculture, Fisheries and Foods, the experiments studied assimilation of C-14 in three crops following atmospheric release to a wind tunnel environment:

- Cabbage;
- Broad beans, and;
- Potatoes (this data was used in the IAEA EMRAS study; [IAEA, 2008]).

The dynamics of C-14 within the crops was also studied. The data collected were intended for use in model testing and included atmospheric C-14 and C-14 in plant components (roots, above-ground plant, tubers, peas, pods). The experimental data for cabbage were used to fit growth curves for the above-ground plant and below-ground plant in SSPAM^{14}C.

Validation of the model was performed by testing it with a decaying source of C-14 to TATM (see Figure 4-8), using the experimental data. As part of this prototype model validation study, parameters, such as those relating to the assimilation following photosynthesis and root uptake in the plant, and respiration from the soil and plant, were tuned to improve the model fit to the empirical plant data.

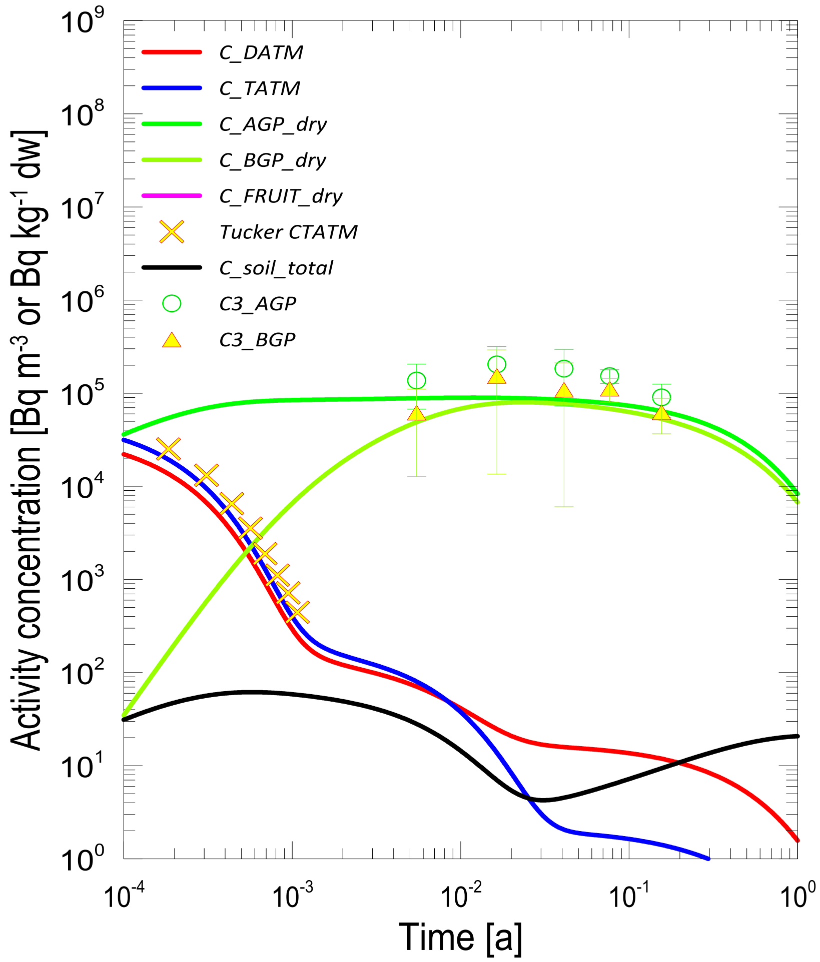


Figure 4-8. Model validation by testing with a decaying source of C-14 to TATM

Without tuning, the model can reproduce activity concentrations in above-ground plant (AGP) but not in below-ground plant (BGP). Tuning of parameters, in this case turning on root uptake or atmospheric pumping, improved the model fit to the BGP data; see Figure 4-8. Unfortunately, no soil measurements were available and hence there is no information to inform the complexity of the soil model and the depth structure, or to inform the tuning of the model. Several alternatives would fit the data.

Testing and validation of this model are described in a report to SSM [Limer et al., 2013a] and in a paper accepted for publication in Radiocarbon [Limer et al., 2013b].

The model SSPAM^{14}C provides SSM with an independent means to assess potential impacts associated with C-14 releases from a range of sources. It was found that one model can be applied to both long-term assessments and short-term data because much of what happens occurs on a timescale of much less than a year (except accumulation of activity in soil organic matter).

Bioprota work has highlighted the importance of considering different ecosystems: crops, grass and natural. The quest for datasets for validation continues, particularly with respect to the soil model.

The discussion identified that it would be worth considering adding the representation of a photosynthate pool in the plant to the model. This pool would be available for plant growth and respiration.

4.7 CIEMAT C-14 MODELLING APPROACH

Danyl Perez-Sanchez (CIEMAT) presented.

The development of the C-14 model started in 2008. It considers the groundwater scenario, where the C-14 initially is introduced into the soil from contaminated irrigation water. Interception by growing crops is ignored and 100% direct deposition to the soil is assumed. Subsequently, a fraction of the C-14 is released by gaseous emission into the atmosphere as CO_2, the remainder leaching to deep soil or lost by erosion. In the atmosphere, CO_2 becomes incorporated into crops via photosynthesis, resulting in increased levels of C-14 in the crops. The uptake of C-14 in crops may also occur via the root system. The CIEMAT C-14 model is based on the RESRAD C-14 model [Yu et al. 2001] applied to Yucca Mountain as a component of the ERMYN Model, since the climate conditions at Yucca Mountain are similar to Spanish climate conditions.

It is a simple model, as shown in Figure 4-9.

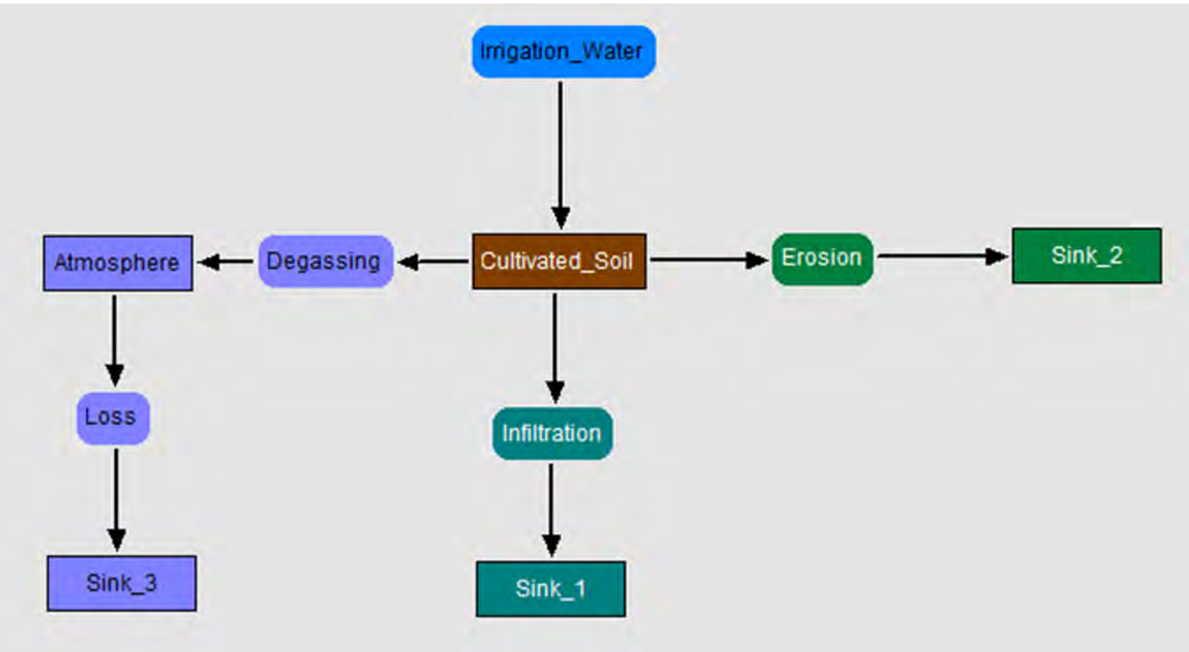


Figure 4-9 The C-14 model developed by CIEMAT

The concentration in the soil (Csoil) is given by:

$$C_{soil} = \frac{C_{wellwater} \cdot IrrigationRate}{(\lambda_{decay} + \lambda_{leaching} + \lambda_{erosion} + \lambda_{evasion}) \cdot \rho_b \cdot d_{soil}}$$

where

- C_{soil} is the activity concentration of C-14 in surface soil for the crop type (Bq kg^{-1});
- $C_{wellwater}$ is the activity concentration of C-14 in irrigation water (Bq m^{-3});

- *IrrigationRate* is the crop irrigation rate for individual crop types, expressed as the average annual irrigation rate (m y^{-1});
- λ_{decay} is the radioactive decay constant for C-14 (y^{-1});
- $\lambda_{leaching}$ is the leaching removal constant for C-14 (y^{-1});
- $\lambda_{erosion}$ is the surface soil erosion removal constant (y^{-1}); and
- $\lambda_{evasion}$,is the emission rate constant of C-14 from the soil to the air (y^{-1}).

Evasion is the dominant removal rate.

The irrigation input is obtained from the difference between the daily crop evapotranspiration, ETcrop, and the effective rainfall, Pe. The parameter values used in the soil model are given in Table 4.5.

Table 4.5. Summary of parameter values used in the model.

Parameter	Description	Value	Unit
Crops Irrigation Rate	Cereal	0,144	m/y
	Root Vegetables	0,18	m/y
	Leafy Vegetables	0,12	m/y
	Fruit	0,084	m/y
Crop Area	A	10000,000	m2
Radioactive Decay	C-14	1,210E-04	/y
Percolation	Cereal	3,037E-01	m/y
	Root Vegetables	1,688E-01	m/y
	Leafy Vegetables	7,320E-02	m/y
	Fruit	1,725E-01	m/y
Leaching removal Constant	Cereal	2,761E-01	/y
	Root Vegetables	1,535E-01	/y
	Leafy Vegetables	6,655E-02	/y
	Fruit	1,568E-01	/y
Erosion rate		0,001	/y
Emission rate of Carbon		14,600	/y
Distribution Coefficient	Kd	0,003	m3/kg
Grain Soil Density		2600,000	kg/m2
Bulk Soil Density		1300,000	kg/m3
Soil Depth	d	0,250	m
Soil Porosity		0,500	-
Moisture Content		0,500	-
Retardation Factor	R	8,800	-

The soil specific activity ranged from 0.6 to 1.3 Bq/kgC for fruit, leafy vegetables, cereals, and root vegetables. The value for cereal was 1 Bq/kgC, corresponding to 3 10^{-2} Bq/kg (f.w.).

C-14 in air is represented by:

$$C_{air} = C_{soil} \cdot \lambda_{evasion} \cdot \rho_b \cdot d_{soil} \cdot \frac{\sqrt{A}}{h_{canopy} \times U_h}$$

Where, C_{air} is the activity concentration of C-14 in the air for the crop type (Bq/m^3), *A* is the surface area of land irrigated with contaminated water, which can be estimated using the volume of contaminated water based on an annual water demand and the average irrigation rate for agricultural land, $\lambda_{evasion}$ is the C-14 emission rate constant of C-14 from the soil to the air (y^{-1}). h_{canopy} is the mixing height of CO_2 (m) and U_h, the annual average wind speed (m/y) at canopy height. The height to which the gaseous C-14 (CO_2) is uniformly mixed, h_{canopy}, depends on the specific application of the parameter depending on the height of type of crops.

The surface layer wind speed profile is given by:

$$u_Z = \frac{u^*}{k} \cdot ln\left(\frac{z}{z_0}\right)$$

This equation can be used to obtain the surface-layer wind profile from the observed wind speed and the aerodynamic surface roughness characteristic of the area of interest. The aerodynamic surface roughness length for the vegetated terrain varies from about 1 cm for short grass to about 10 cm for long grass and crops [Stull, 2001; Sehmel 1984]. The value of *k* (von Karman's constant) is in the range of 0.35 to 0.4 [Stull 2001].The wind profiles are calculated for various values of z_0 and u* (varying *k* does not change wind profiles, because *u** also changes by same factor).

The average wind speed, *U*, in the atmospheric layer limited from the bottom by the surface roughness length, z_0, and from the top by the height of the mixing cell, h_{canopy}, and can be calculated as:

$$\bar{U} = \frac{\frac{u^*}{k} \cdot \left[h_{canopy} \cdot \left(ln\frac{h_{canopy}}{z_0} - 1\right) + z_0\right]}{h_{canopy} - z_0}$$

It must be noted that the gaseous C-14 flux of concern in biosphere modelling originates from irrigated land only. After it is released into the air, C-14 is diluted, mixing with uncontaminated air. The diffusion equation was considered below the height of the canopy. The parameter values are given in Table 4.6.

Table 4.6. Parameter values used in the CIEMAT C-14 model.

Parameter	Description	Value	Unit
Wind Speed	10 m	5,000	m/sec
Crop Heigth	Hmix	0,500	m
von Karman Constant	k	0,400	-
Aerodynamic surface length	Zo=1/3hmix	0,167	m
Friction Velocity	u*	0,488	m/sec
Wind Speed	Crop Height	0,791	m/sec

The resulting specific activity in air is 6.65 Bq m^{-3} for a release of 1 Bq/m^2 to an area growing cereals.

C-14 in crop leaves (produced by photosynthesis) is represented by:

$$C_{Crop,Leaf,C-14} = C_{Air,C-14} \frac{Fa \times fc_{Crop}}{fc_{Air}}$$

Where:

- $C_{Crop,Leaf,C-14}$ is the concentration of C-14 in edible parts of crop type resulting from leaf uptake (Bq/kg wet weight);
- $C_{Air,C-14}$ is the concentration of C-14 in the air for the crop type (Bq/m^3);
- *Fa* is the fraction of air-derived carbon in plants (dimensionless);
- fc_{Crop} is the concentration of stable carbon in the crop (kg carbon/kg wet weight); and
- fc_{Air} is the concentration of stable carbon in the air (kg carbon/m^3).

C-14 uptake by roots is represented by:

$$C_{Crop,Root,C-14} = \frac{C_{Soil,C-14} \times Fs \times fc_{Crop}}{fc_{Soil}}$$

where

- $C_{Crop,Root,C-14}$ is the concentration of C-14 in the edible parts of the crop type resulting from root uptake (Bq/kg wet weight),
- C Soil,C-14 is the concentration of C-14 in surface soil for the crop type (Bq/kg),
- fc_{Crop} is the fraction of stable carbon in the crop type (dimensionless, based on kg carbon/kg wet crop),
- *Fs* is the fraction of soil-derived carbon in plants (dimensionless),
- fc_{Soil} is the fraction of stable carbon in soil (dimensionless, based on kg carbon/kg soil) defined as the mass of carbon per unit mass of soil -this fraction varies depending on soil type, with a typical value on the order of a few percent.

Calculated concentrations in the four crop types showed that 98% of the plant C-14 came from leaf uptake.

The model has been applied to the El Cabril disposal site to update the biosphere safety assessment for LILW, and compare the results with the previous conservative peak effective dose rate of 1E-11 Sv/y from C-14. El Cabril has been in operation since 1992, and the capacity of the centre is envisaged to be sufficient for disposal until about 2020. New cells will be constructed for VLLW. The project has improved and updated the safety assessment modelling for different scenarios (Groundwater and Human Intrusion) and also special radionuclides (C-14, U-238 series), human land use and diets.

5. DISCUSSION OF KEY POINTS ARISING

5.1 PERSPECTIVE ON ISSUES RELEVANT TO THE ASSESSMENT OF RELEASES OF C-14

Tamara L Yankovich presented her perspective on issues relevant to the assessment of releases of C-14.

Nine papers were reviewed, and the background summary, key findings and key discussion points are given below.

5.1.1 S.C. Sheppard, M.I. Sheppard, and F. Siclet, 2006. Parameterization of a Dynamic Specific Activity Model of ^{14}C Transfer from Surface Water-to-Humans

Background Summary

In Sheppard et al. (2006), a model was developed to predict specific activity in water, phytoplankton, fish, crops, meat, milk and air for situations where there are distinct seasonal changes in C-14 activity concentrations in water. The model was designed to estimate the influence of seasonal variations in C-14 activity concentrations on dose estimates for an irrigation-based food chain scenario (i.e., under which isotopic equilibrium could not be predicted).

Data were not available for many of the key parameters required in the model, from the perspective of dose assessment, making it necessary to infer model parameter values. Specifically, data on carbon turnover rates in the various biota compartments were not available (e.g., biochemical turnover, growth dilution, mortality).

Key Findings

To fill in information gaps, values for key parameters were compiled in the paper. These included:

- Isotopic Discrimination Factors (DFs) to reflect natural processes that cause separation of isotopes (e.g., kinetic isotopic fractionation; molecular weight differences);
- First-order rate constants, including: turnover rates within plant and animal tissues (as influenced by growth, mortality, and normal tissue turnover – turnover time determines whether there is net accumulation of C-14 in tissues in a year vs. loss or steady state); and more general rate-limited processes;
- Other ^{14}C-specific parameters, including: water/atmosphere boundary layer thickness (h); soil solid/liquid partition coefficient (Kd); fraction of C-14 added to soil that is Fixed (f_f); canopy dilution factor (CD), or the fraction of C that plants fix from the soil versus the free atmosphere; and
- Concentrations of stable C in various biota and biota tissues (e.g., biological compartments - fats versus carbohydrates versus proteins), where the type of biological compartment can also affect isotopic discrimination, since fats are 'lighter' than carbohydrates (Tagami and Uchida, 2010, see below).

Key Discussion Points

A number of key discussion points were identified regarding the paper. For example:

- What is the relevance to seasonal change in C specific activity as canopy changes energy allocation from foliar growth to tuber growth?

- Application of parameter values, such as Fraction of C-14 Added to Soil that is Fixed (ff) to relative importance of root uptake vs. atmospheric; and

- CD can be especially important in wetlands with groundwater contaminant sources.

5.1.2 B.D. Amiro, Y. Zhuang, and S.C. Sheppard. Relative Importance of Atmospheric and Root Uptake Pathways for 14CO2 Transfer from Contaminated Soil to Plants

Background Summary

Amiro et al. (1991) calculated the relative flux of carbon through roots as a percentage of photosynthesis. Overall, approximately 1-2% of carbon uptake was estimated to occur via root uptake and approximately 1.7% of plant carbon was estimated to be derived from direct uptake via roots in carbonate soils, with much lower root uptake from non-carbonated soils. In addition, the relative flux contributions were estimated as a function of fetch for two crops of different heights. For maize (1.5 m height), a 20-m fetch is required before the atmospheric pathway rivals direct root uptake. At 100-m fetch, the atmospheric pathway is approximately 9-fold more important for beans (0.3 m height) and approximately 4-fold more important for maize (1.5 m height). In general, it was concluded that more exchange with the atmosphere occurs over a larger distance for shorter plants.

Key Findings

Environmental assessment modelling of C-14 requires an estimate of soil-to-plant transfer of contaminants, which can be complex for multiple pathways, especially when source geometry influences the relative importance of pathways. Simple coefficients to estimate transfer from soil to plants (e.g., bulk concentration ratios) are affected by the method used to estimate them. For example, small pot studies can underestimate transfer, and are therefore, not relevant to large, contaminated sites. In addition, a uniform specific activity approach can overestimate transfer for finite areas and is not likely relevant for contaminated soils unless the contaminated area is very large.

Transfer processes and dynamics can be further complicated in natural ecosystems, such as forests or wetlands, which exhibit variability in geometry, moisture conditions, plant assemblages, soil types, redox conditions, the spatial distribution of contaminants and associated pathways (e.g., groundwater-fed versus. atmospheric), and other important factors. Therefore, it is important to determine how we can apply what we have learned from relatively uniform conditions (e.g., crops) to potentially more complex conditions (e.g., wetlands or forests).

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

Background Summary

Carbon-14 was added to the epiliminion of a small, double-basin, 16.1 ha Canadian Shield lake (Lake 226) to investigate primary production and carbon dynamics. In total, 4.6 x 10^{10} Bq of C-14 was added into the epilimnion of Lake 226N and 1.15x10^{10} Bq was added to Lake 226S.

C-14 was primarily lost via $^{14}CO_2$ evasion to the atmosphere, followed by photosynthetic fixation. After this, the C-14 inventory in the epilimnion was controlled by sedimentation of particulate organic C-14 into the metalimnion and hypolimnion.

Experimental data from the study were then used to test 4 models (one simple steady state model; one deterministic model; and two dynamic models).

Key Findings

All models, which included those ranging from relatively simple to highly complex, produced reasonable predictions relative to measured data, taking into account uncertainty. In general, the steady state model was too simplistic to realistically predict water concentrations over the long-term following an acute release, but provided realistic predictions of C-14 retention in sediments. By comparison, more complex, dynamic models are required to predict the internal recycling of contaminants under non-equilibrium conditions.

Key Discussion Points

Based on the findings from the paper, it was determined that it is best to utilize simple models, where possible. This reduces the efforts required in monitoring and model validation, for example, to demonstrate regulatory compliance as part of site licensing, etc. In addition, it is important to select models and modelling approaches that are 'fit for purpose'. For example, key questions could include:

- Is a dynamic model needed for the situation under consideration?
- If not, should a different modelling approach be applied for accidental release scenarios and over what time period?
- Do variations in model predictions fall within natural variability, and if so, can a more simple approach be taken?
- The relative contribution of doses from different pathways may be considered when selecting an appropriate level of modelling effort.

5.1.4 R. Conrad. Soil Microorganisms as Controllers of Atmospheric Trace Gases (H2, CO, CH4, OCS, N2O, and NO)

Background Summary and Key Findings

Trace gas flux between soil and the atmosphere is driven by production, consumption, and transport. Most trace gas production and consumption processes in soil are likely due to microbes, as driven by thresholds (or concentrations below which trace gases are no longer consumed). Conrad (1996) provides a review paper focused on the role of soil microorganisms in cycling of atmospheric gases, including CO, CH_4, etc., and the kinetic characteristics of trace gas consumption by soil, microorganisms, and microbial enzymes.

Trace gases act as substrates for microbe growth and maintenance. Therefore, kinetics of trace gas consumption can be studied to determine whether trace gas metabolism is due to a particular microorganism. Transport processes differ between wetlands and upland soils, due to factors, such as redox conditions, moisture saturation, differences in soil substrates and corresponding microbe composition, and other factors.

Key Discussion Points

Given that kinetics of trace gas consumption can be studied to determine whether trace gas metabolism is due to a particular microorganism, a soil chamber approach, similar to that being at applied of University of Sheffield (see Section 2.1.7) could be considered to gain a better understanding of the influence of microbes on C-14 cycling in soils. . Ultimately, it will be important to identify which processes are more important in driving C-14 flux in wetlands relative to upland environments, and how these drive model uncertainty.

5.1.5 K. Tagami and S. Uchida Estimation of Carbon-14 Transfer from Agricultural Soils to Crops Using Stable Carbon Isotope Ratios

Background Summary

Stable carbon isotopic ratios ($^{13}C/^{12}C$ or $\delta\ ^{13}C$) were used as tracers of carbon transfer from agricultural soils to the edible parts of crop species. Focus was placed on paddy field species (rice) and upland species (leafy vegetables, fruit vegetables, root crops, tubers, legumes, wheat and barley). Study results indicated that carbon transfer factors (TFs) differed between paddy field species (0.11±0.04) and upland field crops (1.0±0.06), and were similar between upland field species. Results concurred with literature values for radishes (0.16 to 1.5).

Signatures of $\delta\ ^{13}C$ were used to discern carbon transport pathways, since isotopic ratios were expected to be constant if all carbon in plants originated from air, whereas changes were expected if there was a contribution from soil.

Key Findings

Assuming a $\delta\ ^{13}C$ of -8 per mil in air and a ^{13}C fractionation ratio of -18 to -20 per mil for photosynthesis by C3-type plants, a $\delta\ ^{13}C$ was estimated, which corresponds to measured data for rice and many upland field crops. Therefore, it was concluded that no soil carbon contribution had occurred. That said, some upland crop species and samples within a given species had $\delta\ ^{13}C$ of less than -29 per mil (i.e., 3/6 tomato samples; 3/5 eggplant samples; 4/7 Japanese radish samples; 2/4 leek samples; and 1 each for cucumber, spinach, *Nozawana*, and white part of leek), possibly because they contained secondary products of the original photosynthesates. A similar concept was discussed by Ryk Klos (see Section 4.6), who described root pumping versus internal plant processes to model root transfer.

Key Discussion Points

Higher carbon transfer factors were observed for upland field crops (1.0) than for paddy field species (0.11), which may be due to the overall drier conditions in upland areas.

Similar patterns were seen in wetlands, where vegetation in saturated zones with clay lenses, etc. showed relatively lower C-14 specific activities than observed in other areas. Further consideration may be required to determine why some individual samples of a given species can be influenced by secondary photosynthates, while other samples of the same species are not.

5.1.6 University of Nottingham (for NDA). Experimental and Modelling Studies of Carbon-14 Behaviour in the Biosphere: Diffusion and Oxidation of Isotopically Labelled Methane (13CH4) in Laboratory Soil Column Experiments

Background Summary

Two laboratory soil column experiments were conducted by the University of Nottingham (U of N) as part of a programme to provide insight into the behaviour and fate of $^{14}CH_4$ following introduction into subsurface soils and its subsequent *in situ* incorporation into vegetation. Specifically, the programme is addressing: 1.) the uncertainty around the potential for $^{14}CH_4$ incorporation into plants, and subsequently, into grazing animals and human consumers; and 2.) the interaction and storage of ^{14}C with soil microbes and in organic matter. The experimental objective is to develop methods to inject and sample small quantities of $^{13}CH_4$ in larger-scale experiments.

Experiments were conducted both in the laboratory and *in situ* at the U of N Experimental Farm, as follows:

- In Experiment 1, loamy sand top soil from the U of N Experimental Farm was packed into columns in the laboratory to a bulk density of ~1.27 g/cm^3 and maintained with a water content of ~20%.
- In Experiment 2, three columns were driven into the soil at the U of N Experimental Farm under undisturbed *in situ* conditions as per laboratory Experiment 1.

Both re-packed and undisturbed soil conditions were tested.

Key Findings

It was determined that two key processes control $^{14}CH_4$ transport and fate in soils – diffusion and oxidation of methane to carbon dioxide by microbes. Re-packed soil showed higher fluxes than undisturbed columns and undisturbed columns were considered more reflective of methane diffusion and oxidation under field conditions.

Key Discussion Points

Based on the work that has been carried out to-date at U of N, a number of key questions have arisen, as follows:

- How do models account for processes that affect disturbance of soils? (e.g., what is the importance of farming practices, such as tilling, on carbon transport?)
- Similarly, are there situations where the re-packed soil experimental study would be relevant in situ?; and
- How will the relative importance of methane diffusion versus oxidation be accounted for in situ?

5.1.7 L. Kumblad, B. Soderback, A. Lofgren, T. Lindborg, E. Wijnbladh, and U. Kautsky. Pools and Fluxes of Organic Matter in a Boreal Landscape: Implications for a Safety Assessment of a Repository for Nuclear Waste

Background Summary

Extensive field studies of different parts of surface ecosystems were carried out at two sites in support of a licence application for a deep repository for spent nuclear fuel. The outputs of detailed carbon dynamic modelling for terrestrial, limnic and marine ecosystems were used to assess major pools and fluxes of organic matter (OM) as a surrogate for radionuclides that have been incorporated in organic matter.

Key Findings

Based on study results, it was determined that the highest proportion of carbon that is incorporated into living tissues occurs in terrestrial catchments. Carbon accumulates in soils and sediments in all ecosystems, with maximum values in shallow, near-shore marine basins. These near-shore basins are considered as focal points for radionuclides at the study site, receiving a relatively large amount of carbon from terrestrial catchments, showing a high net primary productivity (NPP) and high detrital accumulation in the sediments.

Carbon and associated contaminants are primarily transported from marine basins (especially outer basins) to the Baltic Sea through large horizontal water fluxes. In future, near-shore focal points may pose risk of exposure to humans, if they later become agricultural areas due to post-glacial land uplift.

Key Discussion Points

A number of key questions were identified as outcomes of this study. For example:

- Do site-specific conditions influence the application of this modelling approach?
- How well does the use of organic matter pools and fluxes work for different types of radionuclides (e.g., particle-reactive vs. less particle-reactive; volatile vs. not?)
- Landscape changes can result in changes in human exposure scenarios in future. How can these be accounted for?

5.1.8 C. Aulagnier. The TOCATTA-khi Model for Assessing 14C Transfers to Grass: An Evaluation for Atmospheric Operational Releases from Nuclear Facilities

Background Summary

The new version of the TOCATTA radioecological model (Aulagnier et al., in press) can be used to assess C-14 transfer to grassland ecosystems under normal operating conditions for nuclear fuel reprocessing plants. Monthly air, soil, and grass samples were collected *in situ* adjacent to a fuel reprocessing plant. Corresponding meteorological data and hourly atmospheric C-14 activity concentrations (based on Kr-85 measured each minute above the plot) were also collected.

Key Findings

The updated TOCATTA model performs better than the previous version when measured atmospheric C-14 activity concentrations are used as input parameters; otherwise, model uncertainty exists (i.e., when atmospheric C-14 is unknown and must be calculated).

The model adequately predicts C-14 for both intensively managed and poorly managed grasslands, although, an adjustment to the mean C-14 turnover time is required to account for different management practices.

Key Discussion Points

Based on study results, it was determined that land management practices can affect C-14 turnover rates and must be accounted for in models where the distinction between intensively managed and poorly managed grasslands is important. How land management practices are accounted for in the model and the influence of land management on C-14 soil transfer versus photosynthetic uptake should then be considered. The persistence of a C-14 'signal' in the grassland following shut-down periods of the nuclear fuel processing plant should be quantifiedto determine whether it is a transient 'signal' or if it is robust. Finally, the time-frame that is required from a monitoring perspective and a dose perspective is important.

5.1.9 L.M.C. Limer, K. Smith, A. Albrecht, L. Marang, S. Norris, G.M. Smith, M.C. Thorne, and S. Xu. C-14 Long-term Dose Assessment in a Terrestrial Agricultural Ecosystem: FEP Analysis, Scenario Development, and Model Comparison

Background Summary

An international model validation exercise was undertaken within BIOPROTA to compare predicted C-14 dynamics in soil-plant systems and the implications for long-term C-14 dose assessment following release into the biosphere (Limer et al. .2011). Quantitative predictions of C-14 in specific components of dose assessment models were considered, including soil, plant-canopy atmosphere, and plants. The objective of the exercise was to improve confidence in dose assessments for long-term releases of C-14 from radioactive waste repositories into the biosphere.

A Features, Events and Processes (FEP) analysis was undertaken and an interaction matrix developed to detail linkages between FEPs. Available models were then audited against the FEP list. Based on the FEP analysis, a Water Source Interactive Matrix, Gas Source Interactive Matrix, and Soil Layer Interactive Matrix were generated.

Key Findings

Most of the differences between predicted plant C-14 concentrations were due to differences in canopy conceptualization between models. Differences between model predictions seem to be driven by differences in predicted atmospheric C-14 concentrations, which decrease from $\geq$3 orders to a factor of 5 for a given field size, if atmospheric C-14 concentrations are known.

Key drivers that can influence canopy conceptualization include assumptions regarding the degree of mixing of the air used by plants in photosynthesis, degree of openness of the canopy, wind profile both in and above the plant canopy, and interactions between these factors.

A fraction of the C-14 can be stored within the soil subsystem in recalcitrant organic pools that are not readily bioavailable.

Elaborate soil irrigation sub-models do not substantially change model predictions, and simple assumptions that irrigation only depends on yearly average precipitation and evaporation without distinction between plants provides an adequate level of detail in terms of modelling.

Models utilize consistent isotopic ratio approaches, with similar stable C concentrations in air and plants.

Uncertainty related to root uptake or translocation of leaf-deposited bicarbonates does not significantly change model predictions, only accounting up to 2% uncertainty in plant C between models.

Key Discussion Points

Based on the work undertaken as part of this exercise, it was concluded that it is best to keep models simple, where possible. In addition, the report suggests that soil uptake does not substantially affect model predictions; however, during the workshop, there has been significant discussion on the relative importance of soil versus photosynthetic uptake of C-14. Therefore, does it matter? Is it important from the perspective of identifying mechanisms and processes (e.g., seasonal changes in plant allocation from foliar to tuber growth)? The dynamics in the canopy are very important

5.1.10 Potentially Key and Recurring Themes

Through the review of the above documents, a number of key and recurring themes were identified, as follows:

- When should models be kept simple and when should they be more complex? How do time-scales factor in (e.g., application of minute-by-minute Kr-85 measurements to monthly C-14)?
- How conservative is too conservative?

- Biological parameters, such as C turn-over time, etc., can reduce model uncertainty. In such cases, such parameter values may be influenced by site-specific management practices.
- Carbon transport models need to account for different management practices (e.g., ‘intensively managed’ vs. ‘poorly managed’ grasslands; ‘undisturbed’ vs. ‘repacked’ or disturbed soils; etc.).
- Future landscape and corresponding land-use should be considered for long-term, prospective models.
- How do we validate prospective models?
- Different biological compartments can show differences in C contents and isotopic fractionation (e.g., lipids vs. proteins vs. carbohydrates). Does this matter?
- An understanding of carbon pools and their influence on bioavailability requires consideration.
- Fast-growing biota can quickly integrate C-14 into their tissues (e.g., young fish). Biota tissues (e.g., fish) can then serve as long-term C-14 sinks, as can sediments and soils.
- Energetics and energy allocation within organisms, which can change with age, season, and other factors, can influence C-14 cycling. To what degree should this be accounted for?
- What is the importance of time-scale and seasonal processes in C-14 dynamics?
- Differences between model predictions seem to be driven by differences in predicted atmospheric C-14 concentrations in the canopy.
- How relevant are laboratory results in field situations, and how can models account for information gained from the lab for application in situ?
- How relevant are relatively more uniform field situations (e.g., uniform fields) to complex field situations (e.g., canopies; wetlands), and how do models capture this?
- How important is soil uptake from a model prediction point of view? How important from a mechanistic or biological process point of view?
- How important is soil relative to photosynthetic uptake and does it matter?
- How important is soil pathways vs. atmospheric pathways?

Further consideration of the above questions could result in further understanding of C-14 dynamics, which could then be considered in C-14 models.

5.2 ROUND TABLE DISCUSSION

The final session of the meeting was devoted to a round table discussion of key issues of significance that it would be useful to investigate further. These issues are described below and suggestions are made as to the type of investigations that might be undertaken.

It was agreed that data are required relating to the volatilisation of C-14 from soils. This could include flux measurements at the pasture field plots in the vicinity of the Cap de la Hague reprocessing plant. In addition, the current three year RWMD field, laboratory and modelling programme run by the University of Nottingham will be completed in October 2013. This will provide relevant data on losses of methane from the soil following its introduction at a depth of 0.45 m and also on conversion to, and loss of, CO_2. It should be possible to build on this initial programme to undertake detailed kinetic studies of volatilisation in experimentally manipulated soil columns embedded within a realistic field context. The new generation of soil gas samplers used in the phase II field experiment are much more sensitive than those used in phase I, so it may be feasible to sample soil and sub-canopy gas every few minutes and correlate fluctuations in composition with fluctuations in meteoric variables such as barometric pressure. Also, at the Cap de la Hague site, there is the possibility of studying how C-14 concentrations in the pasture plants (grasses) behave when the reprocessing plant is in shutdown mode, as this will be a situation in which the plants are exposed to only very low concentrations of C-14. EdF agreed to look into the possibility of accessing or obtaining relevant data.

It was noted that it would be helpful to distribute the EPRI report on C-14 modelling to BIOPROTA members, but that this is currently not possible because of restrictions imposed by EPRI. It was agreed that EPRI should be contacted directly to see whether arrangements could be made for sharing of the report or data included in it.

In respect of both soil-plant and aquatic system models, there is a need for model results to be confronted with environmental observations. This is required both for refining the models and for building confidence in the results obtained using them.

The soil-plant models developed by Facilia for SKB, by Mike Thorne and Associates for LLWR and by a team of consultants for SSM have similar structures and conceptual bases. Comparisons of the results obtained with these three models would be useful for confidence building, but it should be recognised that there have been extensive exchanges of information between the three modelling teams, and that the three models cannot be regarded as independent. This makes validation against observations of particular importance. The data sets from Cap de la Hague and from the RWMD programme are of potential importance in this context.

The need for a model of C-14 transport in streams and rivers, and of uptake by biota in these environments, was recognised. This is a topic that has been neglected relative to the modelling of C-14 transport in lakes and wetlands. Currently, there is a shortage of data available against which a stream or river model could be validated. However, it is possible that extensive datasets could be obtained from locations upstream and downstream of discharges from nuclear power plants into major European rivers. If so, those datasets would be likely to provide long time series of observations. There is also the possibility of there being datasets related to discharges from hospitals, research centres and radiochemical manufacturers to inland waterways. Studies of the radiological impacts of other radionuclides, such as P-32 and P-33, from such establishments have been conducted, and may provide useful insights relating to development of a C-14 model.

With respect to terrestrial environments, a requirement was identified to be able to define generic models and to determine how to apply those models across different types of environment, e.g. by alternative sets of input parameters.

In the context of wetlands, there was considerable interest in making use of the extensive hydrological and C-14 datasets that are available for Duke Swamp. Because there is an extensive 3D grid of piezometric measurements, it should be possible to construct a physically based hydrological model of the swamp without having sight of the data on measured C-14 concentrations. Indeed, it is possible that hydrogeologists at Chalk River may already have constructed such a model. In the lower levels of the swamp, where there is little biological activity, the C-14 should be transported as a conservative tracer. This can be modelled in physically based surface-water catchment models such as SHETRAN or MIKE-SHE. Furthermore, in such models, representations of biogeochemical cycling can be attached at each node of the modelled 3D grid. Thus, processes resulting in volatilisation of C-14 could be characterised at each of the near-surface nodes, permitting plume depletion to be represented. By this approach, the data from Duke Swamp could be used to achieve a better understanding of the geosphere-biosphere interface.

In view of the usefulness of the Duke Swamp data, which had not been fully appreciated previous to the Workshop, the question was raised as to whether there are other similarly detailed datasets (at Chalk River or elsewhere) that could be studied.

Overall, it was clear that there is the potential to provide a number of extensive and good quality datasets for use in model validation. It was considered that the making of full use of such datasets should take precedence over the acquisition of new datasets. Also, careful thought needs to be given as to how existing datasets should be used. In particular, there is a need to be explicit as to the validation question or questions that are being addressed, the datasets that are relevant to answering those questions, and the ways in which those datasets should be used to maximise their value in respect of key validation issues.

With respect to model development, it was noted that the integration of radio-ecological models is important. In particular, such models need to be scrutinized in a structured way to ensure that no important processes are omitted. A wider range of considerations arise when models are being scrutinized for their fitness-for-purpose for non-human biota assessments than for human assessments, because more pathways of exposure are of potential importance and those pathways are mediated by a wider variety of processes. The area to which a model is to be applied is an important consideration. Requirements may differ for human and non-human biota assessments (e.g. in terms of the resource area to support a critical group relative to the area required to support the local population of a particular species, or the area occupied by a local community or a particular habitat type). In this context, it was noted that a BIOPROTA project has been initiated on spatial and temporal scales of assessment (SPACE).

The discussion of modelling approaches and modelling requirements was considered timely by Posiva, because it will impact the strategy that they will adopted in their next field-sampling campaign.

The importance of biosphere modellers working closely with field investigators was emphasised. In particular, it was considered important that modellers should have direct field experience of the sites that their models are intended to represent.

In respect of the overall assessment process, it was considered that the behaviour of C-14 in the biosphere cannot be an overall ‘show stopper’. If the radiological impact proves problematic, then the source term and transport should first be considered. The biosphere assessment can often be cautious, but the degree of caution adopted needs to be carefully considered and an important aspect of model enhancement and validation is the elimination of highly cautious assumptions made in initial iterations of the assessment process. In the context of model refinement, it can be useful to develop conceptual models at different levels of detail to underpin mathematical model development. Furthermore, it is desirable that the input parameters for a model should be, at least in principle, measurable quantities.

It was noted that the geosphere-biosphere interface for C-14 transport had only been touched on at the meeting, but that this matter will be discussed in more detail in a forthcoming BIOPROTA Workshop on this topic.

Finally, it was noted that a future UK research programme on NORM (funded under the RATE initiative) will be exploring biogeochemical processes in the near-surface zone with a view to generating a nationwide characterisation of the radiation environment. Those BIOPROTA organisations with special interest in impacts of releases of C-14 may find this of interest since it will involve characterisation of the near-surface microbial regime and microbiologically mediated processes.

5.3 SUGGESTIONS FOR FUTURE WORK

The following potential future work activities were identified:

a) The recently developed soil-plant models (e.g. those developed by Facilia for SKB, by Mike Thorne and Associates for LLWR, by IRSN and by a team of consultants for SSM) could be run for well-defined cases and the results compared against each other. The level of agreement could be compared with that obtained in the previous BIOPROTA model inter-comparisons.

b) Ideally, these model comparisons could also be related to specific datasets (potentially those from Cap de la Hague and the University of Nottingham), so that the modelling constitutes a validation test.

c) In the context of wetlands, use could be made of the extensive hydrological and C-14 datasets that are available for Duke Swamp. A 3D catchment model of the swamp could be constructed, calibrated against the hydrological data, combined with a C-14 loss model, and used to predict the 3D distribution of C-14 concentrations in the swamp and the evolution of that distribution with time.

d) Models for C-14 transport in streams, rivers and lakes should be identified (or developed) and a model evaluation and inter-comparison performed. Ideally, these models would be evaluated by comparing the results obtained from them with monitoring data from major European rivers upstream and downstream of the locations of active discharges from nuclear power plants and other types of installation (e.g. hospitals, research installations, radiochemical manufacturers).

e) A review could be conducted to determine general conceptual model structures appropriate to C-14 transport in different ecological contexts. This could then form a basis for evaluating the temporal and spatial scales over which assessments should be undertaken, taking into account the requirement of being able to undertake assessments for both humans and non-human biota. Interactions with the BIOPROTA SPACE project, which has a similar objective but is not limited to C-14, would be beneficial.

f) Those BIOPROTA organisations with special interest in impacts of releases of C-14 should keep up-to-date with a future UK research programme on NORM (funded under the RATE initiative) that will be exploring biogeochemical processes in the near-surface zone with a view to generating a nationwide characterisation of the radiation environment. Although this programme will emphasise NORM, it will be of relevance to C-14, e.g. through characterisation of the near-surface microbial regime and microbiologically mediated processes.

These suggestions are to be explored further as part of the current project. Further information provided by participants after the workshop and by the end of September 2013 is provided in an addendum to this report. Future work will also take into account the results reported in the addendum.

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APPENDIX A. LIST OF PARTICIPANTS

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APPENDIX B. ADDENDUM

This addendum presents further information that became available after the workshop and before the end of September 2013. It presents summaries of two recent papers and recent experimental work.

B1 ADDITIONAL PAPER

Akata N, Abe K, Kakiuchi H, Iyogi T, Shima N & Hisamatsu S, 2013. Radiocarbon concentrations in environmental samples collected near the spent nuclear fuel reprocessing plant at Rokkasho, Aomori, Japan, during test operation using spent nuclear fuel.

Health Physics, 105(3), 236-244.

B1.1 Background Summary

The aim of the study was to determine the contribution of airborne $^{14}CO_2$ discharged from the spent fuel reprocessing facility at Rokkasho to the environmental ^{14}C level within about 8 km of the plant. Discharges from the facility were assumed to occur as $^{14}CO_2$ based on observations of an experimental facility at Tokai, Japan.

The results of monitoring ^{14}C discharges from the reprocessing facility are presented for the period 2006 to 2011. Atmospheric CO_2 and environmental samples were taken from various locations around the site. The main authors are based at the Institute for Environmental Sciences (site ST-1) which is located 2.6 km from the main stack of the reprocessing facility. Environmental samples comprised pasture grass, wild sedge, Japanese radish, cabbage, polished rice and milk. Details of the analytical procedures are given (sampling details, freeze drying, powder production, sample combustion, stable carbon isotope analysis, ^{14}C sample mass spectrometry). The ^{14}C in samples was analysed at the University of Georgia, USA.

B1.2 Key Findings

Elevated ^{14}C in the atmosphere observed during the study was attributed to releases from the facility stack based on a comparison with background levels elsewhere in Japan. The ^{14}C specific activity (Bq g C^{-1}) in samples of Japanese radish, cabbage, polished rice (with the exception of one sample) and milk were not affected by discharges from the facility.

Air monitoring data were compared with results (mBq m^{-3}) from an atmospheric dispersion model (ARAC-2 coupled to a weather model, MM5) that used local meteorological data and stack release data (reported as Bq/month, but based on weekly sampling). Details of the settings used for the weather and dispersion models are given. A detailed comparison of observed $^{14}CO_2$ air concentrations (in excess of background levels) with predicted values is presented for ST-1 along with monthly specific activity (Bq g C^{-1}) data. All environmental samples are reported as a specific activity.

B1.3 Key Discussion Points

The study showed ^{14}C specific activities in wild sedge samples that were slightly above background values and this was particularly true of samples taken east and west of the plant, reflecting seasonal prevailing winds. The only samples to show significantly higher levels (than background) were those of wild sedge and one sample of polished rice from October 2007.

The predicted mean growth period specific activity in atmospheric $^{14}CO_2$ was compared to that measured in wild sedge and showed reasonable agreement. The authors note that the metabolism of carbon was not considered in their simulation, and they suggest a better estimate of plant accumulation might be gained using a metabolic model for carbon assimilation and a detailed source term.

Committed annual effective doses were calculated for inhalation and ingestion pathways. The ingestion dose calculated for rice consumption at the highest recorded specific activity (2007) was 3.6 x 10^{-4} mSv. The peak inhalation dose in 2008 was 7.27 x 10^{-8} mSv.

B2 ADDITIONAL PAPER

Takashi T, Arai R, Nozoe S, Tako Y & Nakamura Y, 2013.A dynamic transfer model for the estimation of ^{14}C radioactivity in Japanese radish (daikon) plants.

Health Physics, 105(2), 121-127.

B2.1 Background Summary

The objective of the study was to develop a dynamic compartmental model of ^{14}C accumulation in Japanese radish (daikon; *Raphanus sativus* L. var. *longipinnatus*) when exposed to temporally changing concentrations of atmospheric $^{14}CO_2$. The crop was selected as one with the highest yield in the area surrounding the Rokkasho spent fuel reprocessing facility which releases ^{14}C to atmosphere. This work follows development of a similar model for potato submitted within the IAEA EMRAS programme. The experimental studies supporting model development used $^{13}CO_2$ as a substitute for $^{14}CO_2$.

The accumulation of ^{13}C is related to specific activity in air and described using parameter R_p which is the ratio of carbon fixed at day *t* after seeding and remaining at harvest to the total carbon in a plant part at harvest. A two-compartment model is presented for which model parameters were obtained from a study of plants at 5 different growth stages exposed to $^{13}CO_2$ for a period of 14 hours. Plants were cultivated using a hydroponic system in a growth chamber and retention of ^{13}C was measured at the time of harvest. All plants were harvested at 49 days, and the period between exposure and harvest ranged from 1 to 31 days.

The dynamic model is described in terms of the rate constants (d^{-1}) determining transfer to 2 compartments (called root and leaf of mass M_R and M_L, respectively) via photosynthesis, a transfer rate between leaf and root (k_T), and loss rates from each compartment due to respiration (k_{LR} and k_{RR}). The root is considered as a single compartment, comprising both the fine inedible root and the enlarged hypocotyl which is edible. The leaves of radish are acknowledged to be the primary carbon receiver, but in this model the root is considered to absorb carbon direct from air to reflect the rapid transfer of carbon from leaves to roots. The redistribution of carbon from leaves to roots is also assumed to occur.

Respiration is considered the sum of two components, maintenance respiration consuming storage substances and growth respiration consuming mainly the currently produced photosynthate. Two variants of the model were compared; in Model-M the respiration rate was considered proportional to the mass of the plant part, and in Model-G the respiration rate was dependent on the growth rate of the plant part. In Model-M the loss rate coefficients (k_{LR} and k_{RR}) were assumed to be constant, and the respiration rate was a product of k_{LR} $M_L(t)$ or k_{RR} $M_R(t)$. In Model-G the loss rate coefficients were calculated form the net carbon gain rate per unit plant mass and a constant derived from the experimental study.

B2.2 Key Findings

The experimental data were used to derive values of $R_L(t)$ and $R_R(t)$ which were then compared with values estimated using the model. Model parameters were determined from the experimental data by least squares fitting. The model outputs and observed data showed good agreement.

B2.3 Key Discussion Points

The model respiration rates were compared with other experimental data and Model-M produced rates comparable to maintenance respiration rates reported elsewhere. However, a large difference was noted in the Model-G respiration rate coefficients between that observed for the root relative to the leaf compartment (the value of $k_{RR}(t)$ was a factor of 30 larger than $k_{LR}(t)$ at harvest). The authors suggest that Model-M gives a better result, stating that maintenance respiration more closely represents the fate of stored carbon.

The authors note that the temperature dependence of respiration rates under field conditions would need to be taken into account if the model was to be used in practice and that model parameters such as mass ($M_R(t)$ and $M_L(t)$) were specific to the growth chamber study.

B3 EXPERIMENTAL WORK

Experimental and Modelling Studies on the Behaviour of C-14 in Agricultural Ecosystems undertaken on behalf of the UK Radioactive Waste Management Directorate over the Last Three Years

B3.1 Introduction

Carbon-14 (half-life 5,730 years) is expected to be released from a geological disposal facility for low- and intermediate-level wastes over a timescale of several thousand years. Both $^{14}CO_2$ and $^{14}CH_4$ will be generated from waste materials within such a facility, but $^{14}CH_4$ is likely to be the dominant carbon-14 species transported in the gas phase, potentially reaching the biosphere at low activity concentrations. Despite much work in recent years on the behaviour of both CH_4 and CO_2 in soils, uncertainty remains over the potential rate of transport of ^{14}C as $^{14}CH_4$ in the soil, its oxidation to $^{14}CO_2$ and subsequent root and foliar uptake by plants.

In view of these uncertainties, the Nuclear Decommissioning Authority Radioactive Waste Management Directorate (NDA/RWMD) commissioned a series of laboratory and field experiments to investigate the transport and retention of $^{13}CH_4$ and $^{14}CH_4$ in agricultural soil. The objectives of the study were:

1. to obtain experimental data on the behaviour of $^{14}CH_4$ and $^{14}CO_2$ in the soil zone, and subsequent uptake of ^{14}C by plants;
2. to interpret the results of the experiments using appropriate models; and
3. to develop an assessment model that can be used to calculate the activity concentrations of ^{14}C in plant material that arise from a below-ground flux of ^{14}C-bearing gas.

This sub-section summarises the key results from the experiments and the associated modelling work. It also summarises the characteristics of the assessment model that has been developed in the light of the work undertaken. The experimental work was carried out by the University of Nottingham, and the modelling work by Amec. The project was overseen by a Steering Group made up of experts from the UK and Canada.

B3.2 Experimental Results

Seven experiments were undertaken: three in the field and four in the laboratory (Atkinson et al., 2011, 2012, In preparation)]. Two of the laboratory experiments used homogenised soil cores, whereas two used undisturbed soil columns taken from the field. Some experiments were vegetated; some were unvegetated. The field experiments used ryegrass in the first year (2011) and spring wheat in the second (2012). The laboratory experiments used a loamy sand soil from the University of Nottingham farm, where the field experiments were undertaken.

Antecedent measurements were taken before methane injection, including various soil properties and soil methane concentrations at a number of depths. Methane fluxes into or out of the soil were estimated using a fixed-volume head space chamber in which the air was continuously stirred. Then methane labelled with ^{13}C was injected at a depth of about 0.50 m, and soil profiles were taken at a number of times following injection. Head-space measurements were again taken at a number of times after injection to estimate fluxes of methane and carbon dioxide.

Here the focus is on the key results from a field experiment (Field Experiment #2) conducted from June to August 2012, and from two soil column experiments (Laboratory Experiments #3 and #4) conducted in the laboratory in 2012 and 2013. These particular experiments have been chosen because, although the earlier experiments measured soil efflux accurately (using the head-space chamber), the soil concentrations were measured less accurately. The later experiments used improved soil gas samplers, which provided better data. Numerical modelling of the earlier experiments suggests that these behaved in a similar way to the later experiments.

Field Experiment #2 investigated the behaviour of discrete pulses of $^{12/13}CH_4$ after injection into the subsoil of a loamy sand at a field site in Nottinghamshire in 2012, which was planted with spring wheat.

Laboratory Experiment #3 investigated the behaviour of a discrete pulse of $^{12/13}CH_4$ after injection into the base of a 0.50 m laboratory column filled with undisturbed soil from the field site. Finally, Laboratory Experiment #4 investigated the behaviour of a discrete pulse of $^{12/13/14}CH_4$ after injection into the base of a 0.50 m laboratory column filled with homogenized topsoil from the field site.

The results show that movement of a discrete pulse of methane through a 0.50 m soil profile is complete within 24 to 48 hours, indicated by a transient increase in soil methane concentrations followed by a return to ambient values. Peak efflux from the soil surface, as measured using headspace chambers, is even more rapid at 3 to 6 hours after gas injection at 0.50 m. Even though the residence time of injected methane within the soil is short, evidence from $\delta^{13}CO_2$[a] measurements indicates that significant oxidation of methane takes place during its diffusive passage through the soil. Modelling studies indicate that this fraction is typically larger than 0.1 and may approach 1.0 (Shaw and Thorne, In preparation; Hoch and Shaw, In preparation).

The majority of the experimental programme has made use of stable isotopic forms of methane ($^{12/13}CH_4$). A laboratory experiment conducted with $^{14}CH_4$ (Laboratory Experiment #4) has indicated that experiments using stable isotopic forms of methane have provided results with direct applicability to the fate and behaviour of $^{14}CH_4$ in the soil-plant-atmosphere system.

B3.3 Wider Context

To complement the experiments, a review of *in situ* studies of methane fluxes across the soil-atmosphere interface in different eco-systems was undertaken (Shaw and Thorne, In preparation). Geometric mean (GM) methane oxidation fluxes from the atmosphere into soils are:

- 1.01 mg m^{-2} d^{-1} for forests (excluding tropical forests) – GM taken over 39 measurements;
- 0.58 mg m^{-2} d^{-1} for grasslands (excluding tropical grasslands) – GM taken over 7 measurements;
- 0.25 mg m^{-2} d^{-1} for arable or agricultural systems – GM taken over 22 measurements.

The methane oxidation fluxes found in the Nottingham laboratory and field experiments (total of 13 measurements) are consistent with the values for arable or agricultural systems given above. These 13 values range from 0.054 to 0.545 mg m^{-2} d^{-1} (Appendix 1 of Shaw and Thorne, In preparation).

In contrast, in wetland environments there is net methane production in the near-surface soils, and thus there is a net flux of methane out of the soils into the free atmosphere.

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[a] This is the deviation of the ^{12}C:^{13}C ratio of a sample (of gas, plant tissue, etc.) from its natural value, and is used as the measure of the quantity of ^{13}C present.

B3.4 Numerical Modelling of the Experiments

The experimental data, mainly the profiles of gas concentrations (i.e. $^{12}CH_4$, $^{13}CH_4$, $^{12}CO_2$ and $^{13}CO_2$) through the soil, but also the effluxes of gases from the soil, provide a consistent picture. Methane ($^{12}CH_4$ and $^{13}CH_4$) diffuses away from the point of injection. As it diffuses, specialist microbes in the soil oxidise the methane to carbon dioxide. In the experiments, the majority of the methane was converted to carbon dioxide. The carbon dioxide ($^{12}CO_2$ and $^{13}CO_2$) then diffuses upwards through the partially saturated soil and into the overlying atmosphere. In order not to stimulate the microbial populations, a rather small volume of methane was injected. As a result, the uptake of labelled carbon dioxide by the plants was below the detection limit.

It follows that the key processes are:

- diffusion of gases through partially saturated soil;
- microbial oxidation of methane; and
- soil respiration, which is the degradation of soil organic matter to produce carbon dioxide; this affects the background concentration profile of CO_2 in the soil.

A sophisticated computational model has been developed that accounts for all of these processes. The model accounts also for isotopic effects; that is, the fact that different isotopic forms of a gas will have slightly different rates for each process.

The model has been applied to interpret the experimental data (Hoch and Shaw, In preparation). The approach was to use the model to simulate Laboratory Experiment #4 first. This particular experiment, which used a repacked, homogenized soil and did not include any vegetation, was the simplest experimental system. Having shown that the model could predict most of the features of this experiment successfully, it was applied next to Laboratory Experiment #3 (intact soil, either with or without vegetation) and finally to Field Experiment #2 (field site with a spring wheat crop).

In applying the model to interpret the experimental data, use was made of existing understanding. For example, the process of gas diffusion through partially saturated soils has been studied extensively. Detailed analyses of a large set of experimental data have led to quantification of the diffusion coefficient as a function of the properties of the soil. The implication for the numerical model is that there is rather little uncertainty in the rates of diffusion of the gases through the soil.

A model in which most of the parameters were based on literature data (i.e. a prediction) provided a satisfactory fit to much of the experimental data. Subsequently, the model was calibrated against the experimental data to improve the quality of the fit.

The combination of the experimental data and the numerical modelling has delivered novel insights into the key process of methane oxidation. In particular, it has been possible to determine the rate at which microbes convert methane to an intermediate form (formaldehyde), and also the fractions of the intermediate that are converted to biomass or carbon dioxide. The experiments that have been undertaken have provided a unique measurement of the rate at which the intermediate breaks down to carbon dioxide.

This sophisticated model has been complemented by a simpler model that was used to analyse the antecedent head space and soil profile measurements, based on assuming a soil column with homogeneous properties and steady-state conditions. The characteristic length scale over which methane will be oxidized in the soil to carbon dioxide is given by $\sqrt{D/(\phi S_g k)}$, where D is the diffusion coefficient of methane in the soil, ϕ is the soil porosity, S_g is the soil gas saturation and k is the first-order rate constant for oxidation of methane. Over the laboratory and field experiments undertaken there have been 13 separate antecedent measurements of the influx of methane. These provide values of k from 5 10^{-5} s^{-1} to 5 10^{-4} s^{-1}, with a geometric mean of 1.8 10^{-4} s^{-1}. The corresponding characteristic length scales are in the range 0.038 to 0.21 m, with an arithmetic average of 0.11 m. The antecedent soil profiles can also be analysed. These provide a good fit to the simple model, and provide vales of k from 7 10^{-6} s^{-1} to 3 10^{-5} s^{-1}, and characteristic length scales in the range 0.20 to 0.46m.

Although this characteristic length scale will be specific to the site and ecosystem under consideration, it seems generally to be of the order of tens of centimetres in agricultural or arable environments. The implication is that most of the radioactive methane migrating from a deep repository is likely to be converted to radioactive carbon dioxide in the soil. There is then the potential for the uptake of radioactive carbon dioxide by plants.

B3.5 Development of an Assessment Model

The potential flux of radioactive methane from a deep repository can be treated as being at quasi-steady state (i.e. the timescale over which the flux of methane varies is much longer than the timescale for methane to diffuse across the near-surface soil). This means that it is justified to use a simple, steady-state diffusion-reaction equation to describe the transport of methane, and its product carbon dioxide, through the soil. In particular, this model depends on two input parameters:

- the diffusion coefficient of a gas through the soil, which in turn depends on the porosity and the gas occupied pore volume in the soil; and
- the rate of methane oxidation.

This model, for a given flux of radioactive methane, provides predictions of:

- the concentration of radioactive carbon dioxide through the soil; and
- the flux of radioactive carbon dioxide from the soil (likely to correspond to almost complete conversion of the methane).

It is considered that much of the radioactive carbon dioxide will migrate upwards from the soil, through the canopy of any overlying vegetation, and into the free atmosphere.

The assessment model uses an atmospheric dispersion calculation to determine the concentration of radioactive carbon dioxide in the atmosphere above the vegetation. Next, the concentration of radioactive carbon dioxide in the canopy of the vegetation is estimated using the concept of an aerodynamic resistance (the difference in the concentration of radioactive carbon dioxide between the plant canopy and the atmosphere is equal to the flux of the gas multiplied by the aerodynamic resistance). Although the aerodynamic resistance has some dependence on plant morphology (e.g. height of plant and density of leaves) and wind category, it is not unduly cautious to use a generic formulation for this quantity. Finally, the concentration of radioactive carbon dioxide in the canopy of the vegetation can be used to determine the specific activity of the canopy atmosphere.

As a result of photosynthesis, the plant will fix carbon dioxide from its canopy atmosphere. Photosynthesis discriminates very slightly (at a level of a few percent) between $^{12}CO_2$ and $^{14}CO_2$. This is not considered to be an important effect, and so the specific activity of the plant will be equal to the specific activity of its canopy atmosphere.

An additional uptake pathway allows for the possibility of ^{14}C being taken up by the plant's roots in the transpiration stream. In this pathway, the concentration of $^{14}CO_2$ in the soil is multiplied by a scaling of the transpiration ratio (i.e. the amount of water taken up by a plant per unit mass of the plant), to calculate an additional source of ^{14}C in the plant. The scaling accounts for the possibility that root uptake of ^{14}C is an active process, and also that ^{14}C will be respired by the roots.

The assessment model assumes that grazing animals will derive most of their food from contaminated vegetation, but humans will derive only part of their diet from the local area (in particular, much of the dietary cereals and sugars would be sourced from elsewhere). It is assumed, cautiously, that at most 30% of carbon in the diet of humans could come from the local area, and so the specific activity of carbon in humans would be diluted from the specific activity of the plants by this fraction.

Finally, a standard calculation is used to relate the specific activity of ^{14}C in humans to the effective dose rate.

The assessment model that is proposed is based extensively on the conceptual understanding that has been developed during previous work, including studies undertaken in the BIOPROTA framework and by LLWR Limited. However, it uses an altered modelling approach, in which concentrations and fluxes of ^{14}C in the system are calculated from a simple representation of the key processes (e.g. molecular diffusion through the soil, methane oxidation by microbes, turbulent diffusion through the plant canopy, and dispersion in the overlying atmosphere).

It is considered that this assessment model has a number of advantages:

- it will be easier to communicate the ideas underlying the model;
- emphasis is placed on the key processes and parameters controlling the uptake of ^{14}C by vegetation, and subsequently by humans; and
- it will be simpler to quantify the key parameters and their uncertainties.

B3.6 Remaining Uncertainties

The remaining uncertainties largely fall into two categories: uncertainties in the canopy and above-canopy model and uncertainties in the soil model. These are considered in turn.

The main uncertainties concern the concentrations of $^{14}CO_2$ in the canopy. These uncertainties arise from the necessary simplifications in the description of the canopy and the atmospheric dispersion model used to describe processes above the canopy. Small plants are assumed, with stable atmospheric conditions and a low wind speed. This is appropriate at the current stage of the programme, but probably constitutes a cautious set of assumptions.

There are fewer uncertainties about the soil model, as it is recommended that conversion of methane should be considered to be complete. Although the model of conversion of CH_4 to CO_2 is physically based, it is a simple representation of a set of complicated processes. However, the diffusion of gases in soils has been extensively studied, and this key aspect of model is believed to be sound.

To date little focus has been placed on how uncertainty should be treated in the assessment model. Although uncertainties in diet are normally neglected, consideration could be given to other uncertainties in the soil, canopy and above-canopy models.

B3.7 References

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The Swedish Radiation Safety Authority has a comprehensive responsibility to ensure that society is safe from the effects of radiation. The Authority works to achieve radiation safety in a number of areas: nuclear power, medical care as well as commercial products and services. The Authority also works to achieve protection from natural radiation and to increase the level of radiation safety internationally.

The Swedish Radiation Safety Authority works proactively and preventively to protect people and the environment from the harmful effects of radiation, now and in the future. The Authority issues regulations and supervises compliance, while also supporting research, providing training and information, and issuing advice. Often, activities involving radiation require licences issued by the Authority. The Swedish Radiation Safety Authority maintains emergency preparedness around the clock with the aim of limiting the aftermath of radiation accidents and the unintentional spreading of radioactive substances. The Authority participates in international co-operation in order to promote radiation safety and finances projects aiming to raise the level of radiation safety in certain Eastern European countries.

The Authority reports to the Ministry of the Environment and has around 315 employees with competencies in the fields of engineering, natural and behavioural sciences, law, economics and communications. We have received quality, environmental and working environment certification.

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