SOIL-PLANT-ATMOSPHERE MODELING IN THE CONTEXT OF RELEASES OF ¹⁴C AS A CONSEQUENCE OF NUCLEAR ACTIVITIES

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ABSTRACT. The need to address radiological impacts from radiocarbon released to the biosphere has been recognized for some time. In 2011, the Swedish Radiation Safety Authority (SSM) commissioned a study to develop a ¹⁴C model of the soil-plant-atmosphere system that would provide them with an independently developed assessment capability. This paper summarizes that study, which comprised a review of contemporary models, the development of a new conceptual model, SSPAM¹⁴C, and the application of SSPAM¹⁴C to a set of experimental data relating to the atmospheric exposure of cabbages.

INTRODUCTION

The need to address radiological impacts from radiocarbon released to the biosphere has been recognized for some time. However, because of its role in biological processes and its ecological cycling, the standard methods employed to model long-term radionuclide transport and accumulation in the biosphere cannot be used satisfactorily for ¹⁴C. The degree of complexity in any ¹⁴C model used must be balanced against the availability of supporting data and the assessment context.

Following Swedish Nuclear Fuel & Waste Management Co.'s (SKB) safety assessment of the lowand intermediate-level waste facility at Forsmark (SFR), the importance of ¹⁴C in long-term dose assessments in Sweden has been re-emphasized (Thomson et al. 2008). The potential impacts associated with ¹⁴C in the biosphere have also been evaluated as being significant in other organizationand country-specific assessments (e.g. Hjerpe et al. 2010), to the extent that the international BIO-PROTA forum has carried out a series of studies of ¹⁴C modeling (Limer et al. 2012). Outcomes from these studies indicate that the choice between dynamic and equilibrium assessment models is of particular interest, raising the issue as to whether dynamic models are useful and/or necessary. Further, the complex processes governing carbon exchanges within the soil and between the soil and the atmosphere are not always well understood, and thus are difficult to represent appropriately in assessment models. Another difficulty surrounding the conceptual model for ¹⁴C concerns the identification of conditions under which mixing and isotopic equilibrium may reasonably be assumed, particularly within the atmosphere.

In this study, a review of existing contemporary ¹⁴C models has been performed, considering models developed to assess operational and accidental releases of ¹⁴C from surface facilities and also those developed for safety assessments associated with the disposal of ¹⁴C-containing radioactive materials. Following on from this review, a model, SSPAM¹⁴C, has been developed and is described herein. SSPAM¹⁴C has been applied to a data set from a study carried out at Imperial College London that involved a series of experiments in which the ¹⁴C activity concentrations in a variety of plants were measured following an atmospheric release of ¹⁴C-labeled carbon dioxide (Tucker and

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Shaw 1997). Some of these data have previously been used in the IAEA's EMRAS program (IAEA 2008). The results of our model review and development study are discussed with a view both to the need for further research and implications for model development.

REVIEW OF EXISTING CONTEMPORARY MODELS

Given the recognized importance of ¹⁴C with respect to its potential impacts within the biosphere, there are already a considerable number of organizations that have developed models to assess these potential impacts associated with a range of nuclear-related activities. The models used by the various organizations fall into 2 broad categories. One subset of the models considered have been developed to consider the potential implications of ¹⁴C releases either through the routine operation of nuclear facilities, or following some form of incident (human or natural). The other subset of models have been developed to consider the implications of ¹⁴C releases following the disposal of ¹⁴C-contaminated wastes, either in near-surface or geological disposal facilities.

Operational and Accidental Release Models

Small amounts of ¹⁴C are generated during the operations of all kinds of nuclear power plants due to capture of neutrons by nitrogen, carbon, or oxygen, present as components of the fuel, moderator, structural hardware, or impurities (NCRP 1985). A fraction of the generated ¹⁴C is released during normal operation of nuclear power plants, mainly in 2 chemical forms: oxidized, i.e. carbon dioxide (CO₂); and reduced, which mostly is in the form of CH₄ (Walker and Otlet 1999). As well as nuclear power plants, ¹⁴C is licensed for use in radiopharmaceuticals and research, and may also enter the surface ecosystem as a result of an accidental release.

Releases from nuclear power plants, routine or accidental, and from other facilities considered in this category, are not continuous because they relate to specific operations or incidents. Consequently, some released radionuclides do not reach equilibrium in the environment. For this reason, some models for routine and accidental atmospheric releases consider processes that might vary over a growing season and, in some instances, a degree of diurnal variation may exist as well.

Waste Disposal Models

The need to address radiological impacts from disposal of radioactive waste containing ¹⁴C has been recognized for some time (e.g. Bush et al. 1984). Particular interest remains in improving the assessment of possible annual individual doses to members of hypothetical exposure groups arising from releases of ¹⁴C to the biosphere from deep and shallow radioactive waste disposal facilities, e.g. the Swedish SFR facility (Thomson et al. 2008), the UK's surface low-level radioactive waste disposal facility (Limer et al. 2011a,b) and site-generic models for a variety of waste types, including ion exchange resins/process water (Magnusson et al. 2008) and graphite (Limer et al. 2010).

Model Comparison

The models considered in this review are summarized in Table 1, with the key observations from the review summarized in Table 2. Irrespective of the source term, a specific activity approach is used to determine the plant ¹⁴C concentration. In a specific activity approach, it is assumed that the ¹⁴C reaches equilibrium in some components of the system in the same proportions with stable carbon. The movement of ¹⁴C and stable carbon is then treated dynamically between some model compartments, and not others.

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$${}^{14}C_{plant} = {}^{14}C_{plant\ environment} \cdot \left(\frac{{}^{stable}C_{plant}}{{}^{stable}C_{plant\ environment}}\right)$$
(1)

Typically the soil, if explicitly represented, and atmospheric compartments are modeled dynamically. In the majority of models developed for application to short-term releases, the plant is also given a degree of dynamism by considering isotopic dilution due to plant growth (e.g. Sheppard et al. 2006a; Keum et al. 2008).

Model type	Agency/Organization	Model name(s)	References
Operational and accidental	UK Food Standards Agency	STAR, PRISM, sewage sludge	Smith et al. 1994; Maul et al. 2005; Thorne et al. 2003
release	Studsvik	POM ¹⁴ C	Aquilonius and Hallberg 2005
	Canadian Standards Agency	N288.1	CSA 2008
	Électricité de France	OURSON	Sheppard et al. 2006a,b
	Korea Atomic Energy Research Institute	Unnamed	Keum et al. 2008
	Japanese collaboration	Unnamed	Tani et al. 2011
	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)	TOCATTA	Aulagnier et al. 2012; Le Dizès et al. 2012
Waste disposal	UK Nuclear Decommissioning Authority Radioactive Waste Management Directorate	Enhanced RIMERS	Thorne 2005; Thorne and MacKenzie 2005
	SKB and Posiva	Unnamed	Avila and Pröhl 2008
	Agence nationale pour la gestion des déchets radioactifs	AquaC_14	Albrecht 2010; Albrecht and Miquel 2010
	Low Level Waste Repository Ltd	Thorne-Limer	Limer et al. 2011a,b

Table 1 Models considered in the qualitative review.

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Aspect of model	Operational models	Waste disposal models
Time steps	Often subannual	Equilibrium conditions or annual average assumed
Source	Gas from above ground Irrigation water Short-term, episodic	Gas from below ground Upwelling water Irrigation water Long-term
Soil	Not always explicitly modeled. One or multiple compartments.	
Plant	Often multiple compartments Dynamic plant growth Isotopic dilution due to new growth	Typically a single compartment Static plant biomass No isotopic dilution
Atmosphere	Sometimes multiple compartments	
Plant ¹⁴ C concentration	Specific activity approach (pho- tosynthesis)	Specific activity approach (photosynthesis) Sometimes root uptake

In instances where the source term is ¹⁴C-labeled gas, consideration needs to be given as to whether the gas is CO_2 or CH_4 . Whereas CO_2 is readily available for plant uptake (via photosynthesis), CH_4 needs to be oxidized to CO_2 before it is available to the plants (e.g. Le Mer and Roger 2001). The degree of oxidation will depend upon the soil microbial population present. Any ¹⁴CH₄ that is oxidized, either in the atmosphere, soil, or water, is available for subsequent uptake and assimilation by plants as ¹⁴CO₂. ¹⁴C in plants can then be ingested and transferred to animals and humans. Where

multiple plant compartments are used, then consideration must be given to which compartments are ingested by animals and humans, as these compartments may differ in both stable and ¹⁴C concentrations.

Although there is some limited evidence to suggest that up to a few percent of a plants carbon might result from direct uptake by roots (Vourinen et al. 1989; Sheppard et al. 1991), it is generally considered that photosynthesis is the dominant, if not only, means by which plants obtain carbon. Given this, it is necessary to consider the profile of photosynthesis through the plant canopy as well as that of the profile of ${}^{14}CO_2$. There is an argument that the profile of the uptake of carbon is dependent upon the canopy density and the penetration of light through the canopy (Figure 1; Monsi and Saeki 2005). Further, as indicated in Figure 1, it is possible that the greater plant mass of photosynthetic issue is found in the upper part of the plant canopy, particularly for a broadleaf plant (Figure 1a).



Figure 1 Productive structure of some plant communities. The dashed thick line shows the relative light intensity (*I*, %). (A) Broadleaf type: *Chenopodium album var. centrorubrum*-consociation, measured on 28 June 1949. (B) Grass type: *Pennisetum japonicum*-consociation (with fruits), measured on 28 September 1949. F = Fresh weight of the photosynthetic tissue in g per 50×50 cm². C = fresh weight of the non-photosynthetic tissue in g per the same area. SN = stem number in 50×50 cm². Reproduced from Monsi and Saeki (2005).

For a release of ¹⁴C from an aboveground source, it might be argued that the profile of ¹⁴C in the plant canopy atmosphere would decrease towards the bottom of the plant. Particularly in the

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instance of a broadleaf plant, this may be a similar pattern as the plant biomass. However, the profile of ¹⁴C in the plant canopy following a release from below ground may not follow the same pattern as the plant biomass. In this instance, it would be reasonable to argue that the ¹⁴C concentration in the canopy air would decrease towards the top of the plant canopy, i.e. potentially behave in an inverse manner to the plant biomass. The implications of assumptions with respect to the plant biomass distribution and the ¹⁴CO₂ distribution in the plant canopy profile in a model that considers the plant and atmosphere each as multiple compartments are discussed further in Limer et al. (2013).

SSPAM¹⁴C: CONCEPTUAL AND MATHEMATICAL MODEL

In order to enable in-depth review of license applications submitted by operators, SSM needs to have independent modeling capacities for assessing the safety associated with releases of radionuclides to the surface environment following the disposal of radioactive waste, operational releases of radionuclides, and incidents or accidents leading to acute releases of radionuclides. If SSM were to use a single model for the assessment of all these release scenarios, then the model must be able to accept a variety of source terms (i.e. gaseous and liquid discharges from above and below ground), and also to consider processes within the ecosystem on a range of timescales. Bearing this in mind, an 11-compartment model (shown in Figure 2) has been developed for SSM. The model, as presently configured, considers each aspect of the soil-plant-atmosphere system by using the maximum number of compartments for each aspect as have been used in pre-existing models. Experimental data may then be used to justify model simplifications in the future.



Figure 2 SSPAM¹⁴C model structure. Two-way exchanges are indicated by thicker arrows.

The model has 6 soil compartments (DPM – decomposable plant material; HUM – humic substances; BIO – biota; INORG – inorganic material; SOL – dissolved C; and SATM – soil atmosphere), 3 plant compartments (AGP – aboveground plant; BGP – belowground plant; and FRUIT – fruit) and 2 atmosphere compartments (DATM – diffusive atmosphere; and TATM – turbulent atmosphere). In

addition, there is also a sink compartment. The sink compartment is used to represent losses of ¹⁴C from the system, including both loss of plant material as a result of harvesting and also ¹⁴C in the turbulent atmosphere carried away from the area of interest by diffusion and air movement. The model is able to accept the whole range of source terms that SSM might need to apply to it. At present, the processes within the model are considered on an annual timescale, making it more suited to the assessment of longer-term releases, though the plant biomass can be static or dynamically modeled, the latter allowing for the potential of isotopic dilution in plant matter due to new growth.

As with the models reviewed, a specific activity approach has been adopted for the uptake of plant ¹⁴C. The plant is assumed to take up 99% of its carbon via photosynthesis, with 1% coming from root uptake. Although the atmosphere is separated into 2 compartments, it is assumed that the aboveground portion of the plant grows inside the diffuse part of the atmosphere only. SSPAM¹⁴C's soil submodel is derived from the RothC model (Coleman and Jenkinson 2005). This model evolved from a model developed to understand carbon turnover in an agricultural soil in the 1970s, using empirical data from extensive field experiments at Rothamsted, UK (Jenkinson and Rayner 1977). Further details of the mathematical model are given in Limer et al. (2013).

Imperial College Experiments

In the 1990s, the UK Ministry of Agriculture, Fisheries and Food (MAFF) funded a study of the assimilation of a selection of radionuclides, including ¹⁴C, following atmospheric discharges. Experiments in a wind tunnel were performed to investigate the uptake of these radionuclides into a range of crop types. For the ¹⁴C experiments, the atmospheric and plant concentrations were measured in 3 crops: cabbage, potatoes, and broad beans. The details of these experiments are given in Tucker and Shaw (1997). The data from the potato experiment was used as input for analyses by the IAEA EMRAS ¹⁴C and tritium working group (IAEA 2008). In the present study, data relating to the cabbage experiment was used to test SSPAM¹⁴C. Figure 3 shows the measured atmospheric profile of injected ¹⁴C and Figure 4, the measured concentrations of ¹⁴C in the various cabbage plant components (leaves, stem, and roots) of one of the replicates (C3). It is the atmospheric profile shown here that SSPAM¹⁴C has been applied to in the following section.



Figure 3 Measured ¹⁴C profile in the atmosphere for the C3 cabbage replicate of the Imperial College experiment.



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Figure 4 Measured ¹⁴C activity concentration in the plant components for the cabbage in C3 replicate exposure (Bq g^{-1}): (a) leaves; (b) stems; and (c) roots. The data points are the means, with the vertical lines representing 1 standard deviation.

RESULTS

In the application of SSPAM¹⁴C to the C3 cabbage data, the area of the model was constrained to that of the wind tunnel, and the concentration of ¹⁴C in the turbulent atmospheric compartment (TATM) was forced to follow a curve fitted to the measured ¹⁴C profile time series (Figure 3). In the initial application, it was assumed that 1% of the plant carbon is obtained from root uptake. As can be seen in Figure 5a, while this led to a reasonable estimation of the aboveground plant ¹⁴C concentration (AGP), SSPAM¹⁴C grossly underestimated the belowground plant ¹⁴C concentrations (BGP). Increasing the proportion of plant C obtained directly from the roots to 10% (as proposed by Livingston and Beall 1934) improves the model fit (Figure 5b), but there is still a significant underestimation as compared the experimental data. Other factors that might affect the calculated BGP ¹⁴C concentration, such as the translocation rate between the AGP and BGP compartments and the respiration rate of the BGP, were considered in additional sensitivity calculations but had a lesser impact than the assumed amount of root uptake of carbon.

DISCUSSION

SSPAM¹⁴C is a model that will provide SSM with an independent means to assess potential impacts associated with ¹⁴C releases to terrestrial environments, from a range of sources. Although the model is still under development, initial tests against experimental data have shown the model is able to recreate aspects of a real system, in particular the aboveground vegetation concentration of ¹⁴C. In the future, consideration may be given as to whether or not the aboveground plant component need be discretized further, and whether the soil aspect of the model could be simplified. However, there is an outstanding need for a comprehensive data set that permits validation of all aspects of the model. Further experimental work, such as the ongoing work funded by the UK Nuclear Decommissioning Authority Radioactive Waste Management Directorate (Atkinson et al. 2011), is anticipated to consider the whole soil-plant-atmosphere system, with a focus on belowground releases of ¹⁴C-labeled





Figure 5 Application of SSPAM¹⁴C to the C3 cabbage experiment replicate. (a) 1% of plant carbon obtained via root uptake. (b) 10% of plant carbon obtained via root uptake. The variation in the measured ¹⁴C concentrations in plant components is shown in Figure 4.

gas. Other organizations, such as IRSN, have also recently undertaken field measurements of ¹⁴C in terrestrial ecosystems following atmospheric discharges (Aulagnier et al. 2012).

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