INTERCOMPARISON OF MODELS OF ^{14}C IN THE BIOSPHERE FOR SOLID RADIOACTIVE WASTE DISPOSAL

Shelly Mobbs[1,2] • George Shaw[3] • Simon Norris[4] • Laura Marang[5] • Trevor Sumerling[6] • Achim Albrecht[7] • Shulan Xu[8] • Mike Thorne[9] • Laura Limer[10] • Karen Smith[1] • Graham Smith[11]

ABSTRACT. Radiocarbon is present in solid radioactive wastes arising from the nuclear power industry, in reactor operating wastes, and in graphite and activated metals that will arise from reactor decommissioning. Its half-life of 5730 yr, among other factors, means that ^{14}C may be released to the biosphere from radioactive waste repositories. These releases may occur as ^{14}C-bearing gases, especially methane, or as aqueous species, and enter the biosphere from below via natural processes or via groundwater pumped from wells. Assessment of radiation doses to humans due to such releases must take account of the major role of carbon in biological processes, requiring specific ^{14}C assessment models to be developed. Therefore, an intercomparison of 5 ^{14}C assessment models was organized by the international collaborative forum, BIOPROTA. The intercomparison identified significantly different results for the activity concentrations in the soil, atmosphere, and plant compartments, based upon the different modeling approaches. The major source of uncertainty was related to the identification of conditions under which mixing occurs and isotopic equilibrium is established. Furthermore, while the assumed release area plays a role in determining the calculated atmospheric ^{14}C concentrations, the openness of the plant canopy and the wind profile in and above the canopy are the key drivers. The intercomparison has aided understanding of the processes involved and helped to identify areas where further research is required to address some of the uncertainties.

INTRODUCTION

The global carbon cycle and the long-term implications of continued radiocarbon discharges from the nuclear fuel cycle have been studied for several decades (Ekdahl et al. 1972; Killough 1980). ^{14}C is present in solid radioactive wastes arising from the nuclear power industry, in reactor operating wastes, and in graphite and activated metals that will arise from reactor decommissioning (Hou 2005, 2007). Its half-life of 5730 yr, among other factors, means that ^{14}C may be released to the biosphere from radioactive waste repositories. The need to address radiological impacts from disposal of radioactive waste containing ^{14}C has been recognized for some time (Bush et al. 1984). These releases may occur as ^{14}C-bearing gases, especially methane, or as aqueous species, and enter the biosphere from below via natural processes or via groundwater pumped from wells. Assessment of radiation doses to humans due to such releases must take account of the major role of carbon in biological processes, requiring specific ^{14}C assessment models to be developed.

There is particular interest in improving the assessment of possible annual individual doses to members of potential exposure groups arising from releases to the biosphere of ^{14}C from deep and shallow radioactive waste disposal facilities, e.g. the Swedish SFR facility (Thomson et al. 2008), and for a variety of waste types, especially reactor operating wastes (Magnusson et al. 2008) and graphite (Limer et al. 2010).

[1]Eden Nuclear and Environment, Eden Conference Barn, Low Moor, Penrith CA10 1XQ, United Kingdom.
[2]Corresponding author. Email: sfm@eden-ne.co.uk.
[3]University of Nottingham, United Kingdom.
[4]Nuclear Decommissioning Authority (NDA), United Kingdom.
[5]Electricité de France (EDF), France.
[6]Low Level Waste Repository Ltd (LLWR), United Kingdom.
[7]L'Agence Nationale pour la gestion des Déchets Radioactifs (ANDRA), France.
[8]Strålsäkerhetsmyndigheten (SSM), Sweden.
[9]Mike Thorne and Associates Ltd, United Kingdom.
[10]Limer Scientific Consulting Ltd, China.
[11]GMS Abingdon Ltd, United Kingdom.

Proceedings of the 21st International Radiocarbon Conference edited by A J T Jull & C Hatté
RADIOCARBON, Vol 55, Nr 2–3, 2013, p 814–825

BIOPROTA (www.bioprota.org) is an international collaboration forum that 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.

Models for ^{14}C behavior in the biosphere were reviewed by BIOPROTA in 2005. That review considered the modeling of releases of ^{14}C both to aquatic (freshwater and marine) and to terrestrial ecosystems. In 2009, BIOPROTA completed a detailed ^{14}C model comparison exercise focusing on terrestrial agricultural ecosystems. This identified significant differences in calculated ^{14}C concentrations in plants arising from different modeling approaches. A follow-up study has now been undertaken and this paper describes the results of the program and the implications for future research. Further details are available in Limer et al. (2012). The overall objective was to improve dose assessments for long-term releases of ^{14}C disposed in radioactive waste.

METHOD

FEP (Features, Events, and Processes) Analysis

The BIOMASS project (IAEA 2003) developed a list of FEPs relating to assessments of the impact of radionuclides in the biosphere. This list was reviewed with respect to the behavior of ^{14}C in the biosphere and a suggested list of FEPs for ^{14}C was developed. The FEP analysis is used to underpin the development of an interaction matrix, which forms the overall conceptual model for the environmental transfer of ^{14}C in the soil-plant system. The 5 models were then audited against this list of FEPs to understand the conceptual differences in the various approaches adopted.

Scenario Development

Three scenarios were selected for the model intercomparison calculations. In all scenarios, it was assumed that the plant obtains 2% of its carbon from the soil and 98% from the atmosphere, via photosynthesis or foliar uptake (Amiro et al. 1991); the crop height was 1 m; and the release area was square with different sizes investigated (1, 10, 100, 1000 m). Climate, soil, and irrigation data were specified.

Groundwater (Irrigation) Source

For the groundwater contamination scenario, it was assumed that ^{14}C in groundwater reaches the plant-soil system via abstraction and irrigation. It was assumed that the activity concentration of the irrigation water is 1 Bq L^{-1} and plant interception of the irrigation water occurs; the stable carbon concentration of the irrigation water is also fixed. The amount of irrigation water and how and when in the season irrigation occurs depend upon the climate, and also on how each individual model considers irrigation (e.g. monthly irrigation events or an annual average). Climate data from an inland site in France were used (Limer et al. 2008) and crop-specific irrigation rates were then determined according to the methodology detailed in Limer et al. (2008). Defining the source water concentration and the irrigation rates means that the calculated soil ^{14}C concentrations are expected to be independent of the assumed irrigated area (the source term has effective units of Bq m^{-2} yr^{-1}).

Gaseous Source

For the gas scenario, it was assumed that ^{14}C-bearing gas (CH_4) enters the soil zone at a rate of 1 Bq m^{-2} yr^{-1}. Reference calculations assumed that the entirety of this flux is oxidized to CO_2 in the rooting zone, thus making it available for plant uptake after degassing. Variant calculations, considering that only part of the ^{14}C-bearing CH_4 reacted, were also performed using some of the models.

Fixed Concentration Assumptions

In addition to the above calculations, a series of calculations were made assuming a fixed concentration of 1 Bq ^{14}C kg^{-1} C in all soil pools, and another series assuming a fixed concentration of 1 Bq kg^{-1} C in the plant canopy atmosphere.

MODEL DESCRIPTIONS

The 5 assessment models developed by the intercomparison members were: AquaC_14, developed for L'Agence Nationale pour la gestion des Déchets Radioactifs (ANDRA), France; SA_Carbon14, developed for Electricité de France (EDF), France; the Avila-Pröhl model, developed for Svensk Kärnbränslehantering AB (SKB), Sweden and Posiva Oy, Finland; SE RIMERS, developed for the Nuclear Decommissioning Authority Radioactive Waste Management Directorate (NDA-RWMD), UK; and the Thorne-Limer model, developed for Low Level Waste Repository Ltd (LLWR), UK. These 5 models are briefly described, paying attention to the conceptual approach to modeling the plant uptake of ^{14}C from the canopy atmosphere. In the majority of the models, the ^{14}C concentration is not explicitly dependent on the stable carbon content since the dependence is implicit in the parameter values selected.

AQUAC_14

This model was originally developed by Penfold and Watkins (1998) for ANDRA and minor modifications were later made (van Hecke 2001; Albrecht 2010). The AquaC_14 model is characterized by a large number of parameters that are specific to carbon, such as stable carbon concentrations in the various compartments and components. The volume concentration in the canopy of plants depends on the concentration of ^{14}C in the soil, the soil density, and the rate of degassing. A geometric factor related to the wind direction, the wind speed, and a parameter relating the size and volume of the canopy ("fetch") are also taken into account. Contamination of plants is assumed to occur via incorporation of ^{14}C as CO_2 during photosynthesis and via root uptake.

SA_CARBON14

This model was developed by EDF (Ciffroy et al. 2005; Sheppard et al. 2006). Input to soil is assumed to be via irrigation with contaminated surface water. The conceptual model for uptake of ^{14}C into crops was adapted during the BIOPROTA exercise and hence 2 approaches were considered: the Sheppard approach and the respiratory recycling approach shown in Figure 1. Partitioning of ^{14}C between labile and recalcitrant forms in the soil is considered. Only a fraction of the CO_2 in the plant canopy is assumed to originate as CO_2 released from the soil. Both approaches are based upon empirical data in which consideration has been given to the crop height, the wind speed, and also the lateral extent of the contaminated area. Absorption of C as CO_2 during photosynthesis is assumed to be the key route of uptake by the plant with other sources assumed to be negligible.

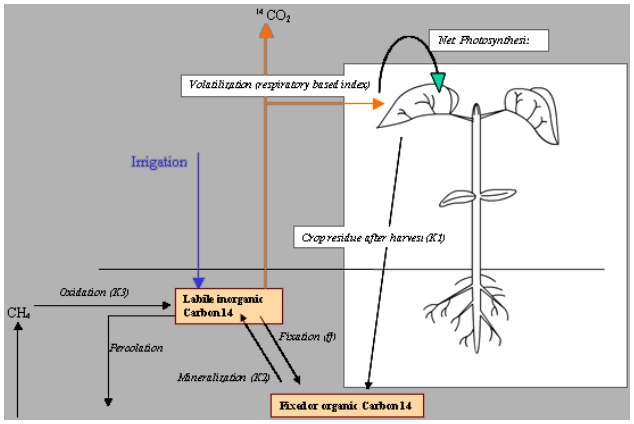

Figure 1 EDF conceptual model for uptake of ^{14}C into plants (respiratory recycling approach).

Simplified Enhanced RIMERS

A simplified version of the enhanced RIMERS model developed by NDA (Thorne 2005, 2006) was used in the intercomparison. This model treats the rapidly exchanging soil solution and soil atmosphere as being in equilibrium and considers only atmospheric transfers, i.e. it neglects the effects of plant uptake in depleting the soil atmosphere and below-canopy atmosphere. It contains 3 compartments: 1) a compartment representing the soil solution and 2) soil atmosphere, which exchanges with 3) a compartment representing the below-canopy atmosphere. This latter compartment exchanges with a compartment representing the above-canopy atmosphere.

The calculated plant canopy ^{14}C concentration depends on air exchange rates between the soil atmosphere, the plant canopy atmosphere, and the above-canopy atmosphere. These exchange rates are calculated based on the wind velocity within the canopy. Clearance of the above-canopy atmosphere is by downwind transport and the ^{14}C concentrations within the canopy are essentially independent of the linear dimensions of the release area.

Avila and Pröhl Model

This model was jointly commissioned by SKB and Posiva in 2008 (Avila and Pröhl 2008). The model assumes that all ^{14}C input into the system with irrigation water is immediately released to the mixing layer where it can be assimilated by the plants via photosynthesis (see Figure 2). The calculated atmospheric ^{14}C concentration depends upon the crop height and the height of the mixing layer, the wind speed at the top of the mixing layer, and also on the vegetation height, the size of the area of consideration, the stable carbon content of the air, and the net primary productivity of the ecosystem.

Thorne-Limer Model

LLWR funded the development of this model (Limer et al. 2011). It considers 2 regions in the aboveground atmosphere: the lower layer only experiences molecular diffusion processes in relation to the movement of molecules of CO_2, whereas the upper layer experiences some degree of turbulent mixing as a result of winds that flow over the area of interest. The thickness of these layers, and the degree of plant uptake of carbon from them, is dependent upon the canopy density, which affects the light intensity and thus the rate of photosynthetic uptake of carbon in the canopy profile. The cal-

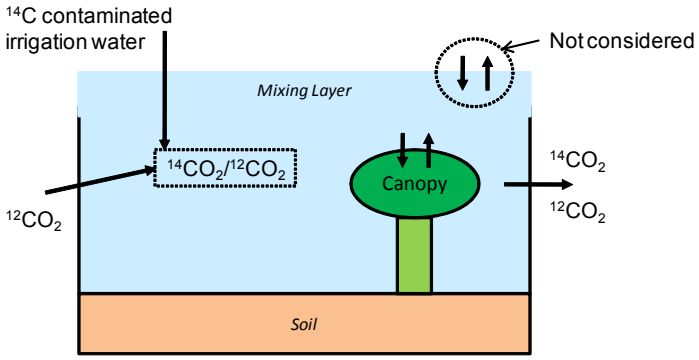

Figure 2 Schematic of the Avila and Pröhl model

culated atmospheric ^{14}C concentration depends upon the crop height, the height of the displacement plane (z_d), and the wind speed. The calculated atmospheric ^{14}C concentrations are independent of the release area, but supplementary calculations with a 2D model demonstrated this to be an accurate approximation.

RESULTS

FEP Analysis and Model Audit

Twelve features were identified in the FEP analysis and used to develop the interaction matrices (IMs) for a water source of ^{14}C and for a gas source of ^{14}C. In addition, an interaction matrix was developed for the soil layer. These IMs were similar to other FEP analyses carried out by members of the IUR Waste Working Group (IUR 2006) and CIEMAT (Aguëro et al. 2006); however, some differences were apparent as a result of the different focus of the work. Our study focused on processes up to and including transport into plants, rather than on dose to humans, and this enabled more detailed breakdown of soil and plant relevant compartments. Specifically, the BIOPROTA IMs divide the canopy atmosphere into 2 compartments, consider above- and belowground plant material as different compartments, and include a "sink" compartment for loss processes. The models were audited against the 12 features to investigate whether they represented the features explicitly (EXP) or implicitly (imp). The results are shown in Table 1.

Table 1 Audit of the 5 models against the 12 features.

Feature	AquaC_14	SA_Carbon14	Enhanced RIMERS	Avila and Pröhl	Thorne-Limer
Source	EXP	EXP	EXP	EXP	EXP
Soil water	imp	EXP	EXP		
Soil solids – recalcitrant		EXP	EXP		
Soil solids – labile		EXP	EXP		
Soil gas	imp	imp	EXP		EXP
Soil microbes			imp		
Mycorrhizae					
Plant canopy atmosphere below z_d	EXP	imp	EXP	imp	EXP
Plant canopy atmosphere above z_d		imp	EXP	imp	EXP
Belowground plant material	imp	imp	imp		imp
Aboveground plant material	EXP	imp	EXP	imp	imp
Sink		EXP	EXP	imp	EXP

CALCULATION RESULTS

The results obtained from each model were compiled and a comparison was made between the calculated ^{14}C concentrations in the various environmental media for the 3 scenarios considered.

Irrigation Scenario

Four of the 5 models were applied to the irrigation scenario, of which 3 reported the calculated ^{14}C concentration in carbon in the soil (Table 2). As would be anticipated given the definition of the source term, the calculated soil ^{14}C concentrations are independent of the irrigated area assumed. Note that the soil ^{14}C concentrations reported from AquaC_14 and SA_Carbon14 relate to soil solids, whereas the soil ^{14}C concentrations reported from the simplified enhanced RIMERS model relate to the soil gas (which is assumed to be in equilibrium with the soil solution). Therefore, only the AquaC_14 and SA_Carbon14 results can be compared, and they agree within a factor of 3.3.

Table 2 Summary of calculated ^{14}C concentrations in soil carbon (Bq kg^{-1} C) for the irrigation scenario

Soil below crop type	AquaC_14	SA_Carbon14	Simplified enhanced RIMERS
Cereals	4.32E-01	Labile: 1.27E0 Fixed: 1.51E-01	4.39E+2
Green leafy vegetables		Labile: 1.39E0 Fixed: 1.51E-01	3.62E+2
Fruit			2.54E+2
Root vegetables			5.44E+2

The calculated atmospheric ^{14}C concentrations vary over 6 orders of magnitude for the smallest irrigated area: the Avila and Pröhl model reporting the lowest values and the EDF model reporting the highest (Table 3).

Table 3 Calculated ^{14}C concentrations in the canopy atmosphere carbon (Bq kg^{-1} C) for the irrigation scenario.

Field length (m)	1	10	100	1000
AquaC_14	4.44E-02	4.44E-01	4.44E+00	4.44E+01
Avila and Pröhl (z_d = 1/6) - cereal	3.46E-04	3.46E-03	3.46E-02	3.44E-01
Avila and Pröhl (z_d = 2/3) - cereal	2.02E-03	2.02E-02	2.02E-01	1.99E+00
Avila and Pröhl (2 m/s wind speed) - cereal	7.57E-04	7.57E-03	7.57E-02	7.52E-01
SA_Carbon14 (Sheppard approach)	—	2.26E+01	5.65E+01	8.48E+01
SA_Carbon14 (Respiratory approach)	—	Cereal: 4.01E+01		
Simplified enhanced RIMERS			Canopy	Above canopy
	Cereal:		4.15E+1	1.63E0
	Green leafy vegetables:		3.46E+1	1.36E0
	Fruit:		2.42E+1	9.50E-1
	Root vegetables:		5.18E+1	2.03E0

However, as the irrigated area considered increases, the difference in the calculated canopy atmosphere ^{14}C concentrations decreases, such that for a characteristic field length of 1000 m, the difference in calculated values is reduced to 2 orders of magnitude. These differences are reflected in the variation in calculated plant ^{14}C concentrations. As an example, the calculated ^{14}C concentrations in cereal for field sizes of length 10 and 1000 m are shown in Figure 3, where z_d is the zero displacement plane expressed as a fraction of plant height and v is the wind velocity at plant height.

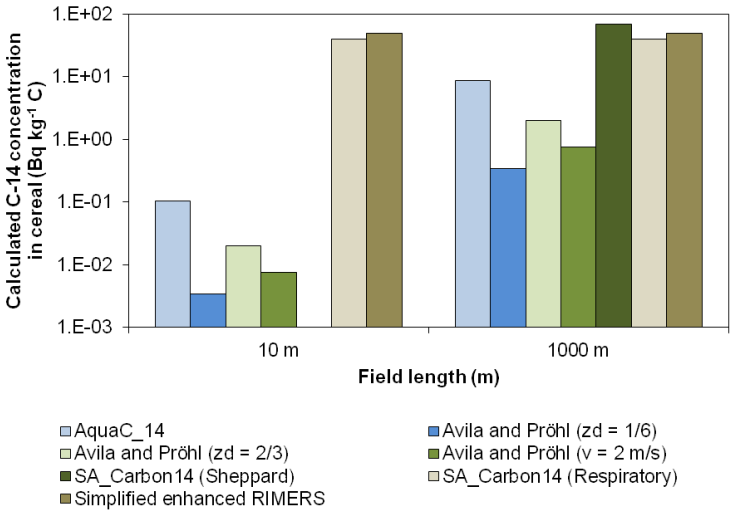

Figure 3 Calculated cereal ^{14}C concentrations for the irrigation scenario; effect of field size

The models with the most cautious assumptions about the plant canopy, e.g. a dense canopy, generate the highest calculated canopy and plant ^{14}C concentrations. However, some of these models appear to be less sensitive to the contaminated area assumed than the models that have a less cautious plant canopy atmosphere approach. This is to be expected. With a dense canopy, the above-canopy wind is not seen within the canopy where movement is largely by vertical diffusion and turbulent mixing.

Gaseous Release Scenario

The calculated soil ^{14}C concentrations, reported by the AquaC_14 and SA_Carbon14 models, are very similar. When the ^{14}C-labeled gas entering the soil is assumed to be 100% CO_2, the calculated soil ^{14}C concentration is of the order 3.9E-03 Bq kg^{-1} C in an available form; SA_Carbon14 also reports 5.7E-04 Bq kg^{-1} C in a more recalcitrant form. The calculated soil ^{14}C concentrations in the Thorne-Limer model are much lower than for the other models; the calculated soil ^{14}C concentration associated with a plant of height 1 m is 2.80E-6 Bq kg^{-1} C. These soil ^{14}C concentrations relate to soil solids. The simplified enhanced RIMERS model reports soil gas ^{14}C concentrations of 3.02 Bq kg^{-1} C.

As with the irrigation scenario, differences between the calculated atmospheric ^{14}C concentrations decrease as the release area increases (Table 4). Thus, for a release area of length 1 m the difference between the models is 5 orders of magnitude, while for an area of length 1000 m the difference drops to 2 orders of magnitude. The differences in the atmospheric concentrations are then reflected in the calculated plant ^{14}C concentrations, as shown in Figure 4 (characteristic lengths of the release area of 10 m and 1000 m are used as examples).

Fixed Concentration Calculations

Fixing the soil ^{14}C concentration in carbon in soil does not reduce the variability in the calculated plant ^{14}C concentrations for areas of a given size (Table 5).

Table 4 Summary of calculated ^{14}C concentrations in the canopy atmosphere carbon (Bq kg^{-1} C) for the gaseous release scenario.

Field length (m)	1	10	100	1000
AquaC_14 (100% CO_2)	3.88E-04	3.88E-03	3.88E-02	3.88E-01
AquaC_14 (11% CO_2)	4.27E-05	4.27E-04	4.27E-03	4.27E-02
Avila and Pröhl (z_d = 1/6)	2.40E-06	2.40E-05	2.40E-04	2.39E-03
Avila and Pröhl (z_d = 2/3)	7.02E-06	7.02E-05	7.01E-04	6.91E-03
Avila and Pröhl (2 m s^{-1} wind speed)	2.63E-06	2.63E-05	2.63E-04	2.61E-03
SA_Carbon14 (Sheppard approach), 100% CO_2	—	7.64E-02	4.27E-01	6.40E-01
SA_Carbon14 (Sheppard approach), 9% CO_2	—	6.88E-03	3.84E-02	5.74E-02
SA_Carbon14 (Respiratory approach), 100% CO_2	—	Cereal (30 m by 30 m field): 3.72E-01 Other crops (10 m by 10 m field): 4.41E-02		
SA_Carbon14 (Respiratory approach), 9% CO_2	—	Cereal: 3.35E-02 Other crops 3.97E-03		
Simplified enhanced RIMERS	Below z_d: 2.88E-01 Above z_d: 1.13E-02			
Thorne-Limer (z_d = 1/6)	Below z_d: 9.45E-01 Above z_d: 3.86E-03			
Thorne-Limer (z_d = 2/3)	Below z_d: 3.77E+00 Above z_d: 3.90E-03			

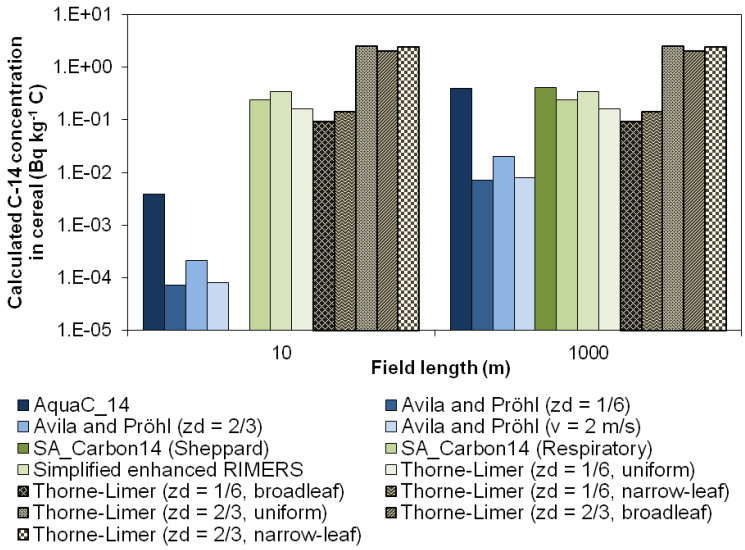

Figure 4 Calculated plant ^{14}C concentrations for the gaseous scenario; release areas with characteristic lengths of 10 and 1000 m.

Table 5 Calculated plant ^{14}C concentrations (Bq kg^{-1} C) when the soil ^{14}C concentration is fixed to 1 Bq kg^{-1} C (10 m characteristic length).

Crop type	AquaC_14	SA_Carbon14 – resp. approach	SA_Carbon14 – Sheppard approach	Simplified enhanced RIMERS	Avila and Pröhl
Cereals	2.20E-01	5.98E+01	—	1.15E-1	1.00E+00
Green leafy vegetables	2.38E-01	7.15E+00	1.24E+01	1.15E-1	1.00E+00
Fruit	2.31E-01	7.10E+00	1.23E+01	1.15E-1	1.00E+00
Root vegetables	2.26E-01	7.08E+00	1.23E+01	1.15E-1	1.00E+00

When the ^{14}C concentration in carbon in the air is fixed, the variability in the calculated plant ^{14}C concentrations drops significantly (Figure 5). This demonstrates that the key processes responsible for model variability in calculated plant ^{14}C concentrations are volatilization and the exchange of gas in the atmosphere and the way they are represented.

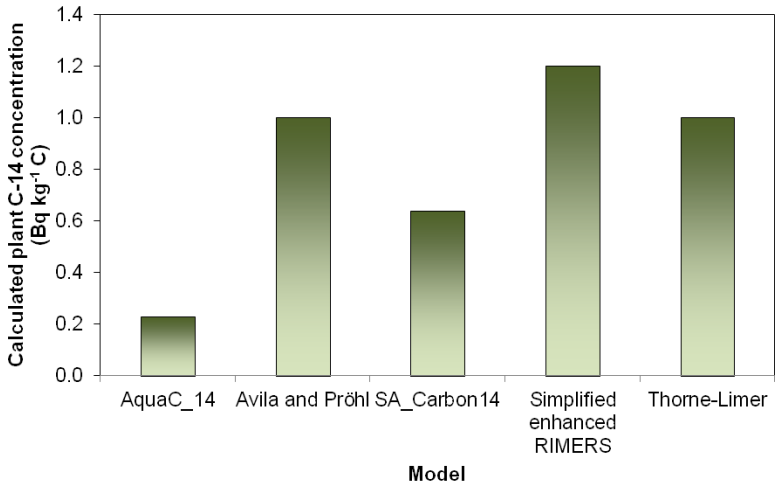

Figure 5 Calculated plant ^{14}C concentrations if the atmosphere is assumed to have a concentration of 1 Bq kg^{-1} C.

DISCUSSION

The FEP analysis, discussion of the models, and examination of results highlight important differences in the conceptual models employed. Their significance in estimating ^{14}C concentrations in different parts of the system is considered in relation to the major model components.

Soil

Within the soil, an important consideration is the form in which ^{14}C is stored, e.g. in readily degradable materials (such as cellulose) and in recalcitrant organic pools (such as humic and fulvic substances) that are not readily bioavailable. Such a distinction is supported by a substantial body of empirical evidence.

Plant Canopy Atmosphere

Following entry to soil (subsurface or irrigation), the differences in the approach to determining the canopy atmosphere concentration of ^{14}C are the major cause of the differences in calculated plant ^{14}C concentrations (Figures 3 and 4). When the atmospheric concentration is fixed, the difference in calculated plant ^{14}C concentration for a given field size dropped from more than 3 orders of magnitude to less than a factor of 5 (Figure 5).

Plant

All models use an isotope ratio approach. Uptake of ^{14}C via root uptake or translocation of leaf-deposited bicarbonates is identified as a minor pathway in all cases and is considered to contribute less than 2% of plant ^{14}C.

Other Factors

The effects of the release area, height of crops, values of z_d, depth of soil layer, etc., on calculated ^{14}C concentrations in the soil, atmosphere, and plants have also been considered. In the AquaC_14, Avila and Pröhl, and SA_Carbon14 models, there is a positive correlation between the size of the release area and the calculated ^{14}C concentrations in each of the model compartments (calculated ^{14}C concentrations in cereal are shown in Figure 6 as an example). These increasing atmospheric and plant ^{14}C concentrations for larger release areas result from the assumption that increasing the release area will increase the time it takes for the contaminated air above the field to exchange with uncontaminated air, and thus the dilution of ^{14}C in the atmosphere is reduced.

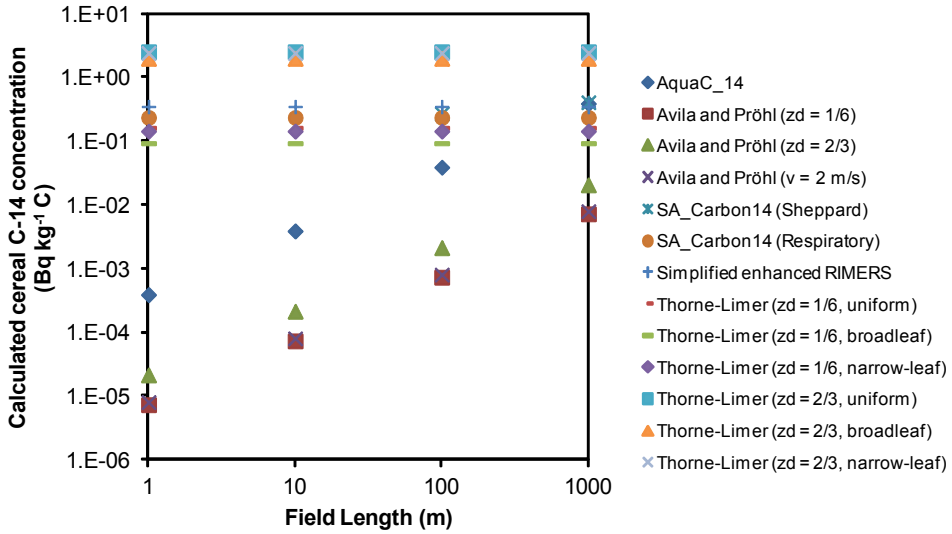

Figure 6 Effect of release area on calculated cereal ^{14}C concentration for the gas release scenario (Bq kg^{-1} C)

Independent of any assumed dependence of the atmospheric ^{14}C concentration on the release area, it is also clear that those models that assume a lesser degree of exchange of the contaminated air in the immediate environment of the plant and the surrounding uncontaminated air will naturally calculate higher plant ^{14}C concentrations for a given ^{14}C flux entering the system.

CONCLUSIONS

The results show the importance of conceptualizing the dynamics of ^{14}C (and stable C) within the plant canopy atmosphere. Models in which the canopy air (main source of C for photosynthesis) is assumed to be subject to a relatively small degree of mixing yield higher calculated plant ^{14}C concentrations than models assuming a greater degree of mixing with uncontaminated air. While the assumed release area plays a role in determining the calculated atmospheric ^{14}C concentrations, it is the assumed degree of openness of the canopy and the wind profile both in and above the plant canopy that are the key drivers in determining the concentration of ^{14}C in the atmosphere used by the plant for photosynthesis. Furthermore, there is an interaction between these factors, with release area being of greater importance for a well-ventilated, open plant canopy.

This study does not imply that any particular approach is "right" or "wrong." Rather, the information should be used to develop a consensus on processes that should/should not be considered in models

for ^{14}C transfer in the biosphere for radioactive waste disposal. Clearly, the type of agricultural practices (such as intense fertilization yielding larger crops and closed canopies) plays a major role, but cannot be predicted for the long term. It is therefore reasonable to consider the potential for exposure based on knowledge of present-day conditions at different sites and in different conditions.

Experimental work is underway, including a 3-yr NDA RWMD-funded program to further develop understanding of ^{14}C behavior in soil-plant systems in field conditions through the introduction of ^{13}C-labeled methane (as a non-radioactive surrogate for ^{14}C) into subsurface soil to determine uptake to vegetation. Results will help to reduce those uncertainties having the most influence in the behavior of ^{14}C in agricultural ecosystems and will support future safety assessment calculations relating to ^{14}C. Future considerations for research may include forms of carbon other than methane (i.e. dissolved organic substances) entering soil, and uptake via aquatic pathways.

ACKNOWLEDGMENTS

This study was supported financially by the following organizations: ANDRA; EDF; LLW Repository Ltd (UK); the National Cooperative for the Disposal of Radioactive Waste (Nagra, Switzerland); the Nuclear Decommissioning Authority (NDA) Radioactive Waste Management Directorate (RWMD); Nuclear Waste Management Organization of Japan (NUMO); Svensk Kärnbränslehantering AB (SKB, Sweden); and the Swedish Radiation Safety Authority (SSM, Sweden).

REFERENCES

Agüero A, Pérez-Sánchez D, Trueba C, Moraleda M. 2006. Characteristics and behaviour of C-14, Cl-36, Pu-239 and Tc-99 in the biosphere in the context of performance assessments of geological repositories for high-level radioactive wastes. Madrid: CIEMAT.

Albrecht A. 2010. Les équations du modèle de transfert du carbone-14 (^{14}C) dans la biosphère (AquaC_14) et leur intégration dans le code "Modèle Management, MoM"; Note technique. Report number: C.NT.ASTR.10.0052.A, ANDRA, Châtenay-Malabry.

Amiro BD, Zhuang Y, Sheppard SC. 1991. Relative importance of atmospheric and root uptake pathways for $^{14}CO_2$ transfer from contaminated soil to plants. *Health Physics* 61(6):825–9.

Avila R, Pröhl G. 2008. Models used in the SFR 1 SAR-08 and KBS-3H safety assessments for calculation of C-14 doses, SKB Rapport R-08-16.

Bush RP, White IF, Smith GM. 1984. Carbon-14 waste management. CEC, Euratom Report EUR-8749.

Ciffroy P, Siclet F, Damois C, Luck M, Duboudin C. 2005. A dynamic model for assessing radiological consequences of routine releases in the Loire River: parameterisation and uncertainty/sensitivity analysis. *Journal of Environmental Radioactivity* 83(1):9–48.

Ekdahl CA, Bacastow R, Keeling DC. 1972. Atmospheric carbon dioxide and radiocarbon in the natural carbon cycle. In: *Brookhaven Symposium on Carbon in the Biosphere*. CONF-720510.

Hou X. 2005. Rapid analysis of ^{14}C and ^{3}H in graphite and concrete for decommissioning of nuclear reactor. *Applied Radiation and Isotopes* 62(6):871–82.

Hou X. 2007. Radiochemical analysis of radionuclides difficult to measure for waste characterization in decommissioning of nuclear facilities. *Journal of Radioanalytical and Nuclear Chemistry* 273(1):43–8.

International Atomic Energy Agency (IAEA). 2003. *"Reference Biospheres" for Solid Radioactive Waste Disposal*. Report of BIOMASS Theme 1 of the BIOsphere Modelling and ASSessment (BIOMASS) Programme, IAEA-BIOMASS-6. Vienna: IAEA.

International Union of Radioecology (IUR). 2006. Recommendations for improving predictions of the long-term environmental behaviour of ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U, Findings of the IUR "Radioecology and Waste" Task Force. IUR Report 6.

Killough GG. 1980. A dynamic model for estimating radiation dose to the world population from releases of ^{14}C to the atmosphere. *Health Physics* 38(3):269–300.

Limer L, Albrecht A, Marang L, Miquel S, Tamponnet C, Nakai K, Gierszewski P, Thorne M, Smith G. 2008. Investigation of Cl-36 behaviour in soils and uptake into crops. A report prepared within the BIOPROTA international cooperation programme and published by ANDRA: C.RO.ASTR.08.0048.

Limer L, Smith G, Thorne M. 2010. Disposal of graphite: a modelling exercise to determine acceptable release rates to the biosphere. A study for the Nuclear Decommissioning Authority, Radioactive Waste Management Division, QRS-1454A-1, Version 2.2.

Limer LMC, Thorne MC, Towler GH. 2011. Assessment calculations for C-14 labelled gas for the LLWR 2011 ESC. Quintessa Limited report to LLWR Limited QRS-1443Z-1, Version 4.0, April 2011.

Limer LMC, Smith K, Albrecht A, Marang L, Norris S, Smith GM, Thorne MC, Xu S. 2012. C-14 long-term dose assessment: data review, scenario development, and model comparison. SSM 2012:47. Available from http://www.stralsakerhetsmyndigheten.se.

Magnusson Å, Strenström K, Aronsson P-O. 2008. ^{14}C in spent ion-exchange resins and process water from nuclear reactors: a method for quantitative determination of organic and inorganic fractions. *Journal of Radioanalytical and Nuclear Chemistry* 275(2):261–73.

Penfold JP, Watkins B. 1998. Transfer of C-14 in the biosphere bibliography and modelling. Rapport QuantiSci n° C NT OQUA 98-001.

Sheppard SC, Ciffroy P, Siclet F, Damois C, Sheppard MI, Stephenson M. 2006. Conceptual approaches for the development of dynamic specific activity models of ^{14}C transfer from surface water to humans. *Journal of Environmental Radioactivity* 87(1):32–51.

Thomson G, Miller A, Smith G, Jackson D. 2008. Radionuclide release calculations for SAR-08. Swedish Nuclear Fuel and Waste Management Company, SKB Report R-08-14. Stockholm.

Thorne MC. 2005. Development of increased understanding of potential radiological impacts of radioactive gases from a deep geological repository: review of FSA and Nirex models and associated scoping calculations. Mike Thorne and Associates Limited report to UK Nirex Limited MTA/P0011b/2005-5: Issue 2, November 2005.

Thorne MC. 2006. Development of increased understanding of potential radiological impacts of radioactive gases from a deep geological repository: sensitivity studies with the Enhanced RIMERS Model. Mike Thorne and Associates Limited Report to Nirex, Report No. MTA/P0011b/2005-10: Issue 2.

van Hecke W. 2001. Intégration du modèle ^{14}C développé par Quantisci au code de calcul Aquabios et comparaison avec le modèle utilisé précédemment. Report ANDRA N° C NT ABSE 00-032/A, Châtenay-Malabry.