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Comparison of modelled uptake to cereal crops of ^{14}C from gaseous or groundwater mediated pathways

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ABSTRACT

Carbon-14 has been identified as one of the more significant radionuclides in solid radioactive wastes in a repository, due to the potential radiological impact arising if ^{14}C were to be released and enter the biosphere. However, the assessment of radiation doses is complicated by the major role of carbon in biological processes, and this has tended to lead to the adoption of a cautious assessment approach.

An international comparison of five models used to predict uptake of ^{14}C to agricultural crops has been undertaken, within the BIOPROTA framework. Processes investigated include conversion of ^{14}C-labelled CH_4 into CO_2 in soils, carbon accumulation in and release from soil carbon pools, gaseous emanation to, and dispersion from, the plant canopy atmosphere and, incorporation into plants by photosynthesis.

For a unit rate of entry of ^{14}C to soil, modelled activity concentrations in cereal crops differ by three to five orders of magnitude. This reflects, in part, differing assumptions for mixing and dispersion of air above the soil surface and within the crop canopy layer. For a unit activity concentration of ^{14}C in air, the modelled uptake to cereal crops converges significantly. Following an assumed irrigation of crops with groundwater containing unit activity of ^{14}C, the predicted uptake to crops varied by two to four orders of magnitude, again largely dominated by assumptions regarding the canopy atmosphere. In all cases, there is some convergence in model predictions as field size increases.

A continuing programme of field research is being undertaken in parallel with the assessment work.

Keywords: disposal, biosphere, dose assessment.

Introduction

Disposal of radioactive waste containing ^{14}C raises issues with regard to the potential generation of $^{14}CO_2$ and $^{14}CH_4$ from wastes containing substantial quantities of degradable organic materials and the need to address radiological impacts from disposal of radioactive waste containing ^{14}C has been recognised for some time (Bush *et al.*, 1983). Across waste management organisations, there is particular interest in improving approaches to 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. van Hecke, 2001; Thomson *et al.*, 2008; Limer *et al.*, 2010). In practice, establishing a disposal system safety case (DSSC) will be a function of the inventory and disposal concept. Just because there is ^{14}C in a disposal inventory and that ^{14}C biosphere studies are being undertaken does not necessarily imply a problem with the safety case. Furthermore, depending on waste

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packaging and repository backfill assumptions, it is anticipated that, in general, CO_2 generated within a repository will be trapped by carbonation of grout in the waste packages or the cementitious back-fill of disposal vaults; however, $^{14}CH_4$ will not be retained and may be released from the near-field, through the far-field (e.g. Harris *et al.*, 2003) with subsequent entry to the biosphere via gaseous or groundwater mediated transfer processes.

Assessment of radiation doses to humans and other biota due to such releases is complicated by to the major role of carbon in biological processes. Large uncertainties in dose assessments have been recognised for some years (e.g. BIOPROTA, 2005), leading to the adoption of cautious assessment approaches, particularly relating to the incorporation of ^{14}C in plants. The Radioactive Waste Management Directorate of the Nuclear Decommissioning Authority (NDA RWMD) and other waste management organizations have cooperated in a programme of model inter-comparison exercises to investigate the significance of different assumptions about ^{14}C behaviour in the biosphere and uptake to plants.

This programme of work has been organized within the BIOPROTA international collaborative forum. A draft report (Limer *et al.*, 2011) and a full description of the work programme is available from the BIOPROTA website (www.bioprota.org). The study has been undertaken to compare and contrast quantitative estimates of modelled ^{14}C concentrations in specific components (soil, plant-canopy atmosphere and plants) of differing dose assessment models representing the soil−plant system. Consideration has been given to the features, events and processes relevant to the behaviour of ^{14}C in the soil−plant system in relation to both gaseous and groundwater mediated transfer processes.

The overall objective of the project is to improve confidence in dose assessments for long-term releases into the biosphere of ^{14}C following disposal in a radioactive waste repository. Results presented here are a subset of the full range of assessments undertaken.

Models

Five models were used in the inter-comparison. Participating models and the organizations that developed them are as follows:

(1) *AquaC_14*, a model developed on behalf of Andra (the National Radioactive Waste Management Agency for France) to describe ^{14}C transport within the biosphere;

(2) *SA_Carbon14*, a specific activity model developed for EDF (the French utility company, Électricité de France) to describe transfer of ^{14}C from surface water to humans;

(3) the "*Avila−Prohl*" specific activity model, developed for SKB (the Swedish Nuclear Fuel and Waste Management Company) and Posiva (the Finnish Nuclear Waste Management Company) for assessment of human exposures resulting from potential underground releases of ^{14}C;

(4) *SE RIMERS*, developed for NDA RWMD (the UK Nuclear Decommissioning Authority Radioactive Waste Management Directorate) to represent the transport of gaseous forms of ^{14}C in the environment;

(5) the '*Thorne−Limer*' model, developed for LLWR (the UK low level waste repository operators) to represent the transport of gaseous forms of ^{14}C in the environment with increased capability to address the exchange of gas in soil−plant−atmosphere.

Andra, 'AquaC_14'

A model for the transfer of ^{14}C in the biosphere was originally developed on behalf of Andra by Penfold and Watkins (1998) and transcribed into the general radionuclide biosphere transfer model, Aquabios, with minor modifications (van Hecke, 2001). The ^{14}C model was later extracted from Aquabios and the current stand-alone version of the model is known as *AquaC_14*.

A detailed analysis of the *AquaC_14* model was carried out in 2010, based on the outcome of the initial ^{14}C inter-comparison, and the slightly updated model has been described by Albrecht and Miquel (2010). Wind speed and fetch can be specified in *AquaC_14* but crop height is not a variable parameter.

EDF, 'SA_Carbon14'

The conceptual model used by EDF is an adaptation of the equations described in Sheppard *et al.* (2006). Input of ^{14}C to soil is from irrigation using contaminated surface water. The model allows for ^{14}C in soil to be partitioned between labile and relatively recalcitrant forms, where the recalcitrant forms may be inorganic (carbonate minerals) or organic (insoluble humic substances). The dynamics of transfer between these fractions is not modelled (i.e. the model simply partitions a portion of the input to recalcitrant or fixed forms).

Carbon dioxide from respiration can be dissipated in two ways: photosynthesis and turbulent mixing with the atmosphere. The *SA_Carbon14* model utilizes the simplifying assumption that only a fraction of CO_2 in the plant canopy originates as CO_2 released from the soil, although recycling of carbon expired as CO_2 and subsequently photosynthetically fixed (expressed as a 'recycling index') can be accommodated.

In *SA_Carbon14*, the absorption of C as CO_2 during photosynthesis is considered as the key route of C uptake by the plant, with all other sources being negligible in comparison. The decomposition of organic matter in the soil, combined with root and soil organism respiration mean that the gas-filled soil pore space is commonly enriched in CO_2 relative to the free above-ground atmosphere. The air in the plant canopy is a mixture of air from the soil pore space and the free atmosphere, and so the plant ^{14}C specific activity is also a mixture of the two sources.

SKB and Posiva, 'Avila–Pröhl' model

In 2008, SKB (the Swedish Nuclear Fuel and Waste Management Company) and Posiva (the Finnish Nuclear Waste Management Company) jointly commissioned a study to develop a set of simplified models for assessment of human exposures resulting from potential underground releases of ^{14}C (Avila and Pröhl, 2008). This study considered models for both terrestrial and aquatic ecosystems. The models are based on a specific activity approach (where specific activity is defined as the activity of ^{14}C per unit mass of total C). As the air in the plant canopy is a mixture of air from the soil pore space and the free atmosphere, the plant ^{14}C specific activity depends on existing ^{14}C in the plant and assumptions relating to the mixing of the air within the plant canopy. Over longer time periods (or at faster plant growth rates) the contribution from the pre-existing plant specific activity lessens compared to uptake from air.

The primary assumption of this model is that all ^{14}C that is input to the system with irrigation water will be immediately released to the mixing layer where it can be assimilated by the irrigated plants via photosynthesis. The model can accommodate differing approaches to setting windspeed and fetch (here fetch defines the average wind travel distance assuming that the release area is circular and is equivalent to the radius of the release area). The approach presented here is based on a windspeed of 5 m s^{-1} at 10 m, with scaling for different values of Z_d (zero plane displacement height) and crop height.

NDA RWMD, 'SE RIMERS'

The NDA RWMD funded the development of the enhanced *RIMERS* model to represent the transport of gaseous forms of ^{14}C in the biosphere (e.g. Thorne, 2006). The model consists of 10 compartments, described as follows. Standing biomass (1) degrades to either decomposable plant material (DPM, 2) or resistant plant material (RPM, 3). Both DPM and RPM degrade by the same pathways, though at different rates, to generate microbial biomass in soil (BIO, 4), physically stabilized organic matter (POM, 5) and chemically stabilised organic matter (COM, 6), as well as CO_2 in soil solution (7). The microbial biomass also respires CO_2, and both physically and chemically stabilized organic matter can eventually degrade to yield CO_2. When CH_4 enters the soil system, it is available to soil microbes and is metabolized by them to CO_2. Carbon dioxide in soil solution (7) exchanges with the soil atmosphere (8), which in turn exchanges with the below-canopy atmosphere (9). Carbon dioxide in the below-canopy atmosphere is available for uptake by plants and incorporation in tissues through photosynthesis or it may exchange with the above-canopy atmosphere (10) and be advected away by the wind.

In this study, a simplified version of the model has been used (*SE RIMERS*), which treats the rapidly exchanging soil solution and soil atmosphere as being in equilibrium and considers only atmospheric transfers; that is, it neglects the effects of plant uptake in depleting the soil atmosphere and below-canopy atmosphere. The *SE RIMERS* model is a 1D model, hence effects of wind fetch are not represented but parameters can be selected to represent wind speed and crop height.

LLWR, 'Thorne–Limer' model

For their 2011 Environmental Safety Case, Low Level Waste Repository Ltd (LLWR) decided to fund the development of a new ^{14}C model, which took into account work within BIOPROTA, to increase the capability to address the exchange of gas in soil–plant–atmosphere, using an approach that includes a more physically based representation of the processes involved (Limer *et al.*, 2011*a*).

This '*Thorne–Limer*' model considers two regions in the above-ground atmosphere. The lower layer only experiences molecular diffusion processes in relation to the movement of molecules of carbon dioxide, whereas the upper layer experiences some degree of turbulent mixing as a result of wind-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. Broad leaf and narrow leaf crop types can be addressed separately, as this affects the degree of light penetration.

The ^{14}C-bearing gas is assumed to enter the soil in the forms of CH_4 and CO_2; CH_4 is assumed to be converted to CO_2 by microbial metabolism in the soil zone. Wind speed and crop height are user defined variables. The approach presented here is based on a wind speed of 5 m s^{-1} at 10 m. Crop height was assumed to be 1 m.

Scenario description

Three scenarios were evaluated, two with release of ^{14}C via gas and one with release via groundwater. Scenario descriptions are intended to be generic and are provided solely for inter-comparison of models, as source terms and transfer mechanisms will vary on a site-specific basis.

Gaseous source

For the gas scenario, it was assumed that ^{14}C as CH_4 enters the soil zone at a rate of 1 Bq m^{-2} y^{-1} and that the entirety of this flux is converted to CO_2 in the rooting zone, thus making it available for plant uptake after degassing. Dispersion of ^{14}C in air, and photosynthetic uptake by plants, was treated in a model-specific fashion.

A second scenario was defined with a fixed concentration of 1 Bq ^{14}C kg^{-1} C in the canopy atmosphere. Dispersion in air was thus not subject to model-specific variability.

Groundwater source

For the groundwater scenario, it was assumed that groundwater contaminated with ^{14}C is abstracted and used to irrigate crops. The concentration of activity in the irrigation water is assumed to be 1 Bq l^{-1} and the stable carbon concentration of the irrigation water is also fixed. The amounts of irrigation water, and how and when in the season irrigation occurs, depend upon the climate. Crop-specific irrigation rates (m y^{-1}) were determined according to the methodology detailed in Limer *et al.* (2008), based on climate data for an inland site in France, defined in a previous BIOPROTA study (Bytwerk *et al.*, 2011). For cereal crops an irrigation rate of 0.144 m y^{-1} was applied.

Depending on modelled parameters, plant uptake may be represented as a result of interception, photosynthesis of volatilized $^{14}CO_2$ and, in solution, via the roots.

Information common to the groundwater and gaseous pathway scenarios

It was assumed that plants obtain 2% of carbon from soil and 98% from atmosphere, via photosynthesis or foliar uptake (Amiro *et al.*, 1991). The stable carbon content (g C kg^{-1} dry weight) of the plants and basis of derivation (e.g. whether generic, crop-specific or edible part-specific values are employed) was defined by each participating organization. A reference crop height of 1 m was assumed. Fixed field sizes were defined. Results are presented here for field lengths of 10 m to 1000 m.

Wind speed is commonly presented for a certain height (e.g. 10 m) and then scaled to the vegetation height. Within the canopy, individual models adopt varying assumptions with respect to the shape of the wind profile. Some consider an average wind speed throughout the entire canopy, others assume the wind speed to be equal to that at the top of the canopy throughout, and some assume a wind speed profile within the canopy which depends upon the location within the canopy. To provide greater commonality in this study a zero plane displacement height, Z_d (m), was defined below which there is zero wind speed. The height of Z_d is expressed as a fraction of the height of the vegetation canopy, Z_c (m). Literature values indicate that Z_d varies from one-tenth to two-thirds of Z_c (e.g. Amiro *et al.*, 1991; Avila and Pröhl, 2008) for a range of vegetation types. In this study, for a wind speed of 5 m s^{-1} at a height of 10 m, a crop height of 1 m, and Z_d of 0.17 m a windspeed at 1 m height of 2.8 m s^{-1} was adopted; for a Z_d of 0.67 m a windspeed at 1 m height of 0.75 m s^{-1} was adopted.

Participants were required to state assumptions adopted to represent different crop types.

Media in which ^{14}C concentrations were calculated

Calculations were undertaken of the concentration of ^{14}C in root vegetables, leafy green vegetables, cereals and fruit (Limer *et al*., 2011*a*). The edible parts of these crops differ (for root vegetables the edible part is below ground; for lettuce and fruit the edible part is above ground; for grain it is only the seed body that is eaten). The focus is on ^{14}C levels in crops because previous studies within the BIOPROTA programme have shown that food-crop ingestion is the dominant exposure pathway for ^{14}C releases and these are the same crops which have been used in previous BIOPROTA studies (Bytwerk *et al.*, 2011; Limer *et al.*, 2008).

Estimates of the concentration of ^{14}C were calculated in crops at the time of harvest together with the ^{14}C concentration in the top soil and plant-canopy atmosphere. Only data for cereals are presented here.

Results and discussion

Results are presented in Fig. 1 for uptake of ^{14}C to cereals for the gas release scenario with ^{14}C as CH_4 entering the soil zone at a rate of 1 Bq m^{-2} y^{-1}. Calculated plant concentrations vary by more than four orders of magnitude at the smallest field length of 10 m. The highest concentrations are predicted by the *Thorne–Limer* model (at ~3 Bq kg^{-1} C, for either narrow leaf (NL) or broad leaf (BL) crops and Z_d set at 0.67 m). The lowest concentrations are predicted by the *Avila–Prohl* model (at ~8×10^{-5} Bq kg^{-1} C for Z_d set at 0.17 m). Both the *AquaC_14* and the *Avila–Prohl* model allow for lateral mixing of air, and consequently are sensitive to field size with loss to adjacent areas.

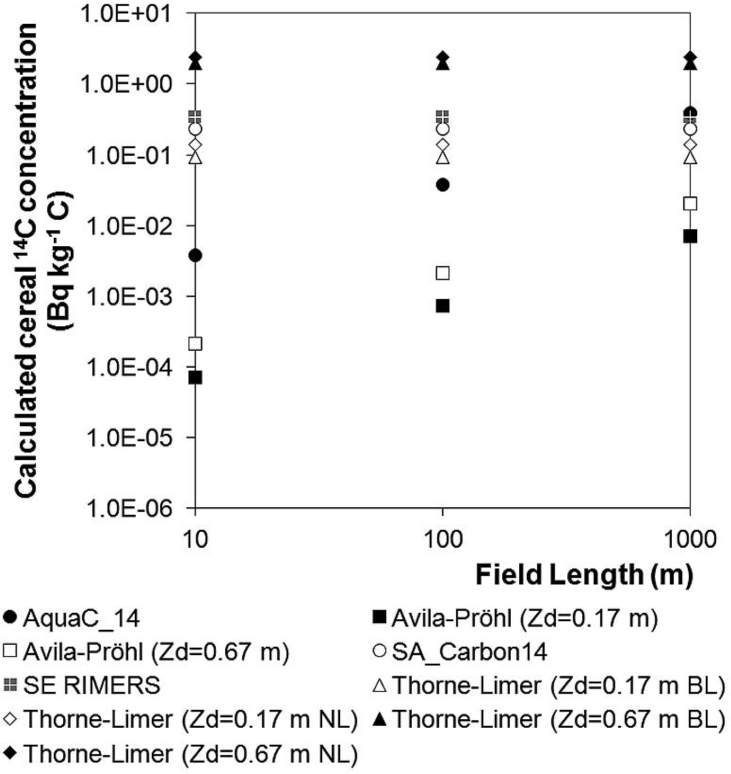

FIG. 1. Calculated cereal ^{14}C concentrations and the effect of field size for the gas release scenario Z_d (for models which incorporate air stratification) is the zero plane displacement height, (m), below which there is zero wind speed; Z_d is expressed here for a crop 1 m high. NL represents 'narrow leaf' crops, BL represents 'broad leaf' crops.

As the field length increases from 10 m to 1000 m, the activity concentration in cereal crops predicted using the *Avila–Prohl* model rises to ~0.008 Bq kg^{-1} C (for Z_d set at 0.17 m). On a nearer like for like basis (i.e. comparing results for the case where Z_d is fixed at 0.17 m for both models) for the 1000 m field length, the *Thorne–Limer* model predicts an activity concentration of about 0.1 Bq kg^{-1} C, indicating greater convergence between the differing approaches. The *AquaC_14*, *SA_CARBON* and *SE RIMERS* models are insensitive to field length or Z_d and predict activity concentrations around 0.4 to 0.6 Bq kg^{-1} C.

If ^{14}C concentration in carbon in the plant air canopy is fixed, at 1 Bq ^{14}C kg^{-1} C, variability in calculated plant ^{14}C concentrations drops to within a factor of around 5 for all models, as indicated in Fig. 2. In the case of the *Thorne–Limer* model and the *Avila–Prohl* models the activity concentration in the cereal is predicted to be 1 Bq ^{14}C kg^{-1} C (i.e. the same specific activity concentration as the air canopy). The *SE RIMERS* model predicts a higher activity concentration (1.2 Bq ^{14}C kg^{-1} C), due to a contribution from root uptake. The *AquaC_14* and *SA_CARBON* models both predict lower activity concentrations (at ~0.2 and 0.6 Bq ^{14}C kg^{-1} C, respectively).

Considerable variability is exhibited in predicted activity concentrations in the cereal crop in the groundwater irrigation scenario (Fig. 3). The dominance of the photosynthetic uptake pathway again means that predicted activity concentrations are sensitive to field size for those models which incorporate lateral mixing of air (*AquaC_14* and the *Avila–Prohl* model). For a field of 10 m length, the lowest predicted crop activity concentration is ~0.005 Bq ^{14}C kg^{-1} C (*Avila–Prohl* model, Z_d set at 0.17 m) and the highest activity concentration is 70 Bq ^{14}C kg^{-1} C (*SE RIMERS*). There is some convergence in predicted activity concentrations as field length increases (with the Avila–Prohl model predicting an activity concentration of 0.5 Bq ^{14}C kg^{-1} C and the *SE RIMERS* prediction being unchanged) although more than two orders of magnitude still separate the highest

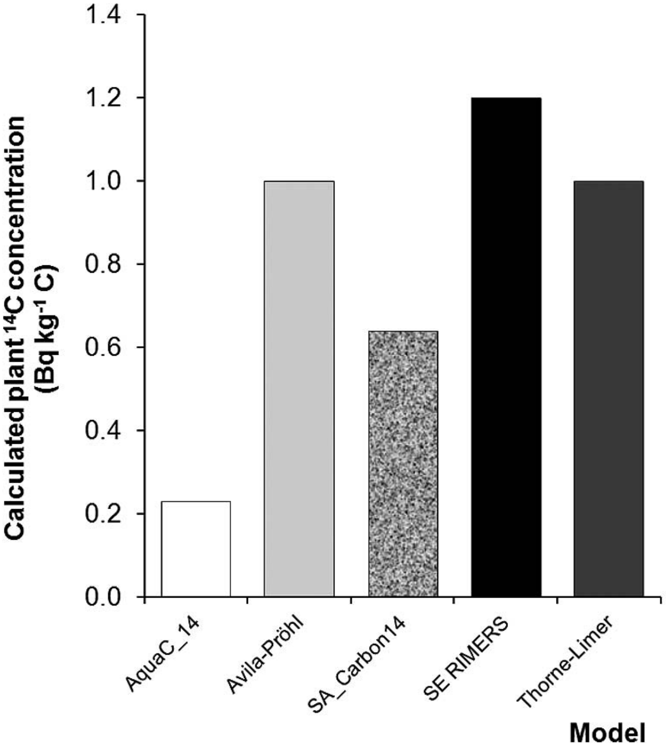

FIG. 2. Calculated plant ^{14}C concentrations assuming an atmospheric concentration of 1 Bq kg^{-1} C.

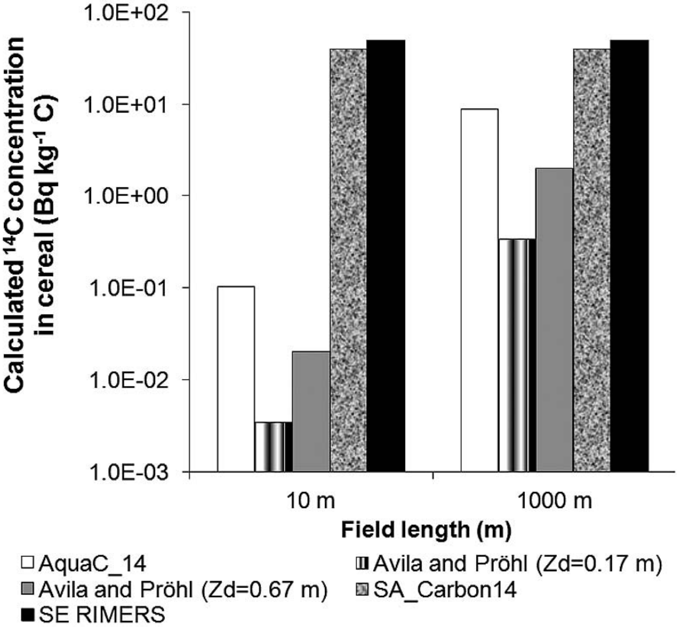

FIG. 3. Calculated cereal ^{14}C concentrations and the effect of field size for the groundwater irrigation scenario Z_d (for models which incorporate air stratification) is the zero plane displacement height, (m), below which there is zero wind speed; Z_d is expressed here for a crop 1 m high.

and lowest results. The differences between the model results for the irrigation scenario reflect both the different approaches to modelling uptake from air and the concentration of ^{14}C labelled CO_2 in air following irrigation with contaminated groundwater. The *Thorne–Limer* model was not applied to the groundwater irrigation scenario.

It is notable that the conceptualization of the canopy atmosphere varies between the models used in this study, and this is the cause of the majority of the variability in calculated plant ^{14}C concentrations for both the direct gas release and groundwater irrigation scenarios. Looking at the two extremes from Fig. 1, the calculated plant ^{14}C concentrations in both the *Avila–Pröhl* and *Thorne–Limer* models are dependent upon the plant height and the relative height of Z_d with respect to the plant height. In the *Avila–Pröhl* model, the correlation between plant height and calculated plant ^{14}C concentration is negative (i.e. increasing crop height is associated with decreasing activity concentration), whereas for the *Thorne–Limer* model it is positive (i.e. increasing crop height is associated with increasing activity concentration). In the *Avila–Pröhl* model, increasing the plant height lowers the calculated ratio of $^{14}C/^{12}C$ in the atmosphere. In the *Thorne–Limer* model, increasing the plant height automatically increases the thickness of the compartment of air which sees only diffusive air mixing (increasing the calculated ^{14}C concentration in the lower atmosphere compartment) and decreases the thickness of the compartment which is subject to turbulent mixing.

The variability in model outputs does not imply that the approach taken by any contributory organisation is ‘right’ or ‘wrong’. Rather, the study output provides information with respect to the dynamics of the models currently used by various waste management organizations. Nonetheless, it is noted that the approach of models in which the air the plant uses for photosynthesis is assumed to be subject to a relatively small degree of mixing, naturally leads to higher calculated plant ^{14}C concentrations than in the approach adopted by other models in which the air the plant uses is subject to a greater degree of mixing. Although the assumed field size (and thus fetch) plays a role in determining the calculated atmospheric ^{14}C concentrations, for those models which incorporate lateral air movement, in reality it is the assumed degree of openness of the canopy and the wind profile both within and above the plant canopy which are more likely to be the key drivers in determining the ^{14}C concentration in the CO_2 that the plants absorb for photosynthesis. In this context it is noted, based on unpublished information, that LLWR is currently considering revisions to the *Thorne–Limer* model to introduce turbulent mixing of air throughout the crop canopy, with air flow determined from a resistance model to create a profile through the crop stand.

Continuing research programme

A three-year NDA RWMD funded experimental programme is underway to further develop an understanding of the transport of ^{14}C in soil–plant systems in field conditions (Atkinson *et al.*, 2012). The parameter to which the models are most sensitive is the assumed mixing of air within the plant canopy and between adjacent air layers; and (for smaller field sizes) lateral air movement. The behaviour and fate of ^{14}C-labelled methane introduced into subsurface soil and subsequent incorporation into vegetation is being determined experimentally using ^{13}C as a non-radioactive surrogate for ^{14}C. Isotope ratio mass spectroscopy is used to determine $^{13}C/^{12}C$ ratios. Currently, small-scale laboratory investigations are being undertaken. These experiments will provide guidance and confidence in the design of the field experiments. Results will provide information that will help to further bound the uncertainties regarded as having the most influence in the behaviour of ^{14}C in agricultural ecosystems, and as such support future safety assessment calculations relating to ^{14}C.

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