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Consideration of canopy structure in modelling ^{14}C-labelled gas behaviour in the biosphere for human dose assessments

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Abstract. The LLW Repository Limited has recognised the potential importance of the processes being considered in the BIOPROTA ^{14}C working group and funded the development a new ^{14}C model that addresses the exchange of gas in a soil-plant-atmosphere system. This model considers two regions in the above-ground atmosphere and utilises concepts from the field of micrometeorology to describe the exchange of air between these regions and losses from the area of interest. 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 which flow over the area of interest. The thicknesses of these layers depend upon the canopy density, which will affect the light intensity and thus the rate of photosynthetic uptake of carbon in the canopy profile. Model results demonstrate the impacts of ^{14}C-labelled gas from the soil upon the calculated ^{14}C concentration in plants for a variety of plant species (pasture and garden crops) and subsequent doses to human exposure groups. The technical modelling work described has been funded by the LLW Repository Ltd in support of its 2011 Environmental Safety Case.

1. INTRODUCTION

The global carbon cycle and the long-term implications of continued ^{14}C discharges from the nuclear fuel cycle have been studied for several decades. Gaseous release of ^{14}C from the geosphere to the biosphere is primarily of interest in the context of disposal of low and intermediate level radioactive wastes containing substantial quantities of degradable organic materials. These wastes can give rise to releases of CO_2 and CH_4 labelled with ^{14}C. In addition, in anaerobic conditions, bulk hydrogen can arise from metal corrosion and can act as a carrier gas. With inorganic wastes, e.g. graphite and metal wastes, bulk CO_2 and CH_4 gas production may be of much less significance. When assessing the potential impact of gaseous release of ^{14}C-labelled gas from a repository to the biosphere much can be learnt by considering the impacts of atmospheric releases of ^{14}C-labelled gas, e.g. from a nuclear power plant. For example, the impacts of atmospheric releases of ^{14}C-labelled CO_2 upon potatoes and rice were considered by the International Atomic Energy Agency's (IAEA) EMRAS tritium/^{14}C working group [1, 2].

The Low Level Waste Repository (LLWR) Limited Site Licence Company undertook a programme of work that resulted in the production of an Environmental Safety Case (ESC) which was submitted to the Environment Agency (EA) in May 2011. An important component of the arguments presented were calculations of the long-term radiological impact from disposed wastes. ^{14}C represents a key contaminant for a number of the exposure pathways explored in these analyses. Of these the gas pathway is of particular importance. The assessment of the ^{14}C-labelled gas pathway comprised four stages: firstly consideration was given to the near field processes that would influence the temporal and spatial release of ^{14}C-labelled gas, including its chemical speciation [3]; secondly, consideration was given to the transport of bulk gas through the engineered cap for the facility; thirdly, an evaluation of the

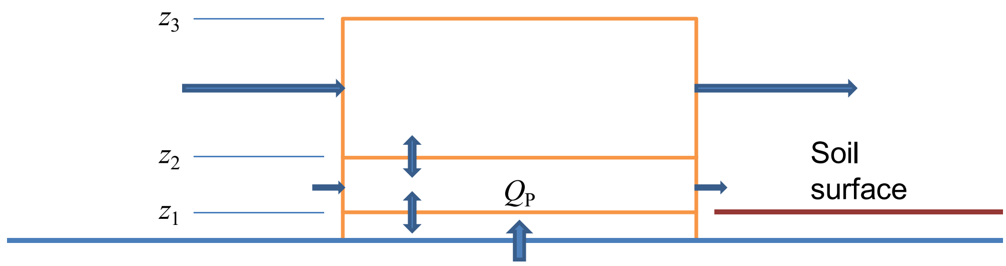

Figure 1. Vertical structure of the plant canopy atmosphere model.

behaviour of ^{14}C in the biosphere. In the final stage the preceding analyses were brought together to address assessment specific questions. This assessment, undertaken by Quintessa Ltd, is presented in full in Limer et al. [4]. In this paper, the ^{14}C soil-plant model developed specifically for the 2011 ESC is described, together with the calculated plant ^{14}C concentrations resulting from a flux of 1 Bq m^{-2} y^{1}. The calculated values are discussed in the context of the BIOPROTA ^{14}C working group activities [5].

2. MATHEMATICAL MODEL

2.1 Definition of plant available carbon

The CH_4 fluxes must be converted to CO_2 fluxes by microbial metabolism in the soil zone if they are to be available directly to plants. The degree of metabolism may be affected by the mass flux of CH_4. The total plant available flux of ^{14}C-labelled CO_2, Q_P (Bq m^{-2} s^{-1}) is given by:

$$Q_P = Q_c + \mu \cdot Q_m \tag{1}$$

Here the subscripts c and m correspond to CO_2 and CH_4 respectively. The conversion efficiency μ lies in the range [0, 1] and is determined by the flow of CH_4 per unit area. In the reference calculations presented in the 2011 ESC it is cautiously assumed that 100% of any CH_4 released is fully oxidised to CO_2 and is thus available for plant uptake.

2.2 Transport in the plant canopy

From the studies undertaken in support of BIOPROTA using the enhanced RIMERS model [6], it was shown that a single-pass model is appropriate to representing the flow of ^{14}C-labelled CO_2 through the plant canopy and its uptake in photosynthesis. The geometry for the model used in the 2011 ESC is shown in Figure 1.

The vertical structure shown in Figure 1 distinguishes the base of the model from the soil surface. Thus, the region [0, z_1] corresponds to soil solution plus soil atmosphere. The height of this compartment corresponds to the thickness of the soil plus subsoil layer of the repository cap. The height of the second layer is chosen to be equal to the height where turbulent mixing in the plant canopy commences, and thus is plant-type specific. Layer 3 contains the top of the plant canopy; its height is fixed for all plant types to ensure that there is a minimum of a 10 m thickness of free air that overlies the canopy.

Vertical air exchanges can occur between the layers and also a horizontal flow of air can occur both within the upper part of the plant canopy and above the canopy air (these are in the third compartment). To make the formulation of the model identical for each layer, such a flow is included in the model formulation, though it would be usual to set the value to zero for the lower canopy and soil zone compartments. It is noted that the movement of ^{14}C-labelled CO_2 through the canopy will be fairly rapid. Therefore, variations in wind direction do not need to be considered.

Computationally, the model is fully defined by the following ordinary differential equations. Note that these equations are specified only for ^{14}C. For the lowest layer, the governing equation is:

$$w \cdot X \cdot \theta \cdot \frac{dC_1}{dt} = Q_P - U_1 \cdot z_1 \cdot w \cdot C_1 + w \cdot X \cdot V_{2,1} \cdot C_2 - w \cdot X \cdot V_{1,2} \cdot C_2 \quad (2)$$

where X (m) is the lateral extent of the model cell and w (m) is its width.

The left hand side corresponds to the rate of change of the activity content of the layer, i.e. C_1 (Bq m^{-3}) is the activity concentration in the soil atmosphere and θ(-) is the porosity of the soil (the volumetric concentrations in soil atmosphere and soil solution are taken as identical, but different concentrations can be accommodated by defining an effective porosity). The second term on the right corresponds to advective horizontal transport out of the layer, where U(m s^{-1}) is the wind velocity. The final two terms on the right correspond to vertical transport into and out of the layer from the overlying layer. This is represented by the concept of an effective velocity V(m s^{-1}), though, in practice, the process is treated diffusively. The relevant equations for layers 2 and 3 are:

$$w \cdot X \cdot (z_2 - z_1) \cdot \frac{dC_2}{dt} = -\left[U_2 \cdot (z_2 - z_1) + X \cdot V_{2,1} + X \cdot V_{2,3}\right] \cdot w \cdot C_2 + w \cdot X \cdot V_{1,2} \cdot C_1 + w \cdot X \cdot V_{3,2} \cdot C_3 \quad (3)$$

$$w \cdot X \cdot (z_3 - z_2) \cdot \frac{dC_3}{dt} = -U_3 \cdot (z_3 - z_2) \cdot w \cdot C_3 + wX \cdot V_{2,3} \cdot C_2 - w \cdot X \cdot V_{3,2} \cdot C_3 \quad (4)$$

All the terms in these equations are well-defined, except for V and U. These are discussed below.

2.2.1 Representation of Vertical Air Flow

Transport vertically in and through the canopy is treated as a diffusion-like process, where the diffusion coefficient D(m^2 s^{-1}) is represented by:

$$\begin{aligned} D &= D_{\mathrm{air}} && z \le z_{\mathrm{d}} \\ &= D_{\mathrm{air}} + k\, u_* (z - z_{\mathrm{d}}) && z > z_{\mathrm{d}} \end{aligned} \quad (5)$$

where k is von Karman's constant [7], u_* is the friction velocity that depends on the wind speed away from the surface and the surface roughness length [8], which is in turn related to the height of the roughness elements (here the plant canopy), z is the height above the surface (determined by the compartment geometry), z_{d} is the height above the surface where the wind speed is taken to fall to zero (the zero displacement plane), and D_{air} is the diffusion coefficient in air [9]. The zero displacement plane is assumed to lie within the canopy of the 'dominant' plant species that determines the wind profile close to the surface, i.e. $z_{\mathrm{d}} < z_{\mathrm{C}}$, where z_{c} (m) is the canopy height. Allen et al. [10] observed that for a wide range of crops the zero plane displacement height, z_{d} (m), can be estimated from the canopy height, z_{C} (m), by assuming that it is two-thirds of the canopy height. A study of barley in Estonia concluded that the calculated z_{d} could be taken as three-quarters and two-thirds of z_{C} for a dense and a moderate canopy respectively [11]. In some literature sources, z_{d} is given as a sixth or tenth of the vegetation height (e.g. [12]).

There is no unique representation of the transport resistance between layers j and j + 1 and the associated diffusion-like transfer rate between compartments. Here, an expression is used for the transport resistance that takes account of variations in the diffusion coefficient in the donor and receptor compartments in the direction of transport.

$$\Omega^{j,j+1} = \frac{1}{2}\int_{\varsigma+h^j}^{\varsigma+h^{j+1}} \frac{dz}{D(z)} \tag{6}$$

Here $\Omega^{j,j+1}$ (s m^{-1}) is diffusive transport resistance, ζ represents the position of the interface, and h (m) is the length of the compartment in the direction of transport. As wX_j is the area between the compartments, the rate constant for flow between them is $1/h^j\Omega^{j,j+1}$ s^{-1}. This rate constant can also be written as $wX_jV_{j,j+1}/wX_jh^j = V_{j,j+1}/h^j$. Thus:

$$V_{j,j+1} = \frac{1}{\Omega^{j,j+1}} \tag{7}$$

2.2.2 Representation of horizontal velocity

Within the plant canopy, it is appropriate to take $U = u*$ above the zero displacement plane and to set it to zero below the zero displacement plane. However, above the canopy the air flow regime is different. Here, it is assumed that the wind speed at 10 m, U_{10} (m s^{-1}), is provided as input. The wind speed is matched to u$_*$ at the height of the canopy z_c using:

$$\begin{aligned} U_j &= U_{10}\left(\frac{u_*}{U_{10}}\right)^{(10-z)/(10-z_C)} && \text{with } z = z_j \qquad \text{for } z_j > z_C \\ &= u_* && \text{for } z_d < z_j \leq z_C \\ &= 0 && \text{for } z_j \leq z_d \end{aligned} \tag{8}$$

Full details of the parameterisation of the equations relating to the transport of gas in the model compartments are given in [4].

2.3 Representation of plant uptake

The equations given in above allow the time-dependent concentrations of ^{14}C in air to be calculated. These values are expressed for the soil atmosphere/soil solution (C_1) or for the above-soil atmosphere (C_2 and C_3) in units of Bq m^{-3}. As described in detail in [4], it is possible to convert these volumetric ^{14}C concentrations to specific activities, $S_{,j}$ (Bq kg^{-1}[C]). Assuming that plants obtain fractions g$_j$ of their carbon from the various layers j, the specific activity of plant carbon from that area, S_P (Bq kg^{-1}[C]), is given by:

$$S_P = \sum_j g_j S_j \tag{9}$$

When defining how much carbon the plant takes up from the various compartments, one possible assumption is that the plant takes up carbon uniformly, e.g. if the plant was 1 m high then the plant would take up 1/100$^{\text{th}}$ of its carbon per cm height. However, it is accepted that the dominant means of carbon uptake by the plant is via photosynthesis, i.e. that root uptake can be neglected, and that photosynthesis does not occur at a uniform rate through the plant height. One particular factor which will influence C uptake in the canopy is the light intensity. It is possible to used Beer's Law to represent the light extinction curve in the canopy as an exponential function [13, 14]:

$$I = I_0 \cdot \exp(-K \cdot LAI) \tag{10}$$

Here I is the shaded light intensity under the leaf area index *LAI*, I_0 is the original incoming light intensity (i.e. the light intensity at the top of the canopy), and K is the extinction coefficient.

Experimental data indicated that the extinction coefficients K of the majority of plant communities fall into two groups, with the light extinction by a given leaf layer being somewhat faster in broad-leaf types than in grass-types [13, 14].

Anten [15] proposed that the light saturated rate of leaf photosynthesis through a plant canopy, P, would follow a similar curve:

$$P = P_0 \cdot \exp\left(-\frac{K_N}{K_{df}} \cdot K_{df} \cdot LAI\right) \tag{11}$$

Here P_0 is the photosynthetic uptake at the top of the canopy, K_N is the coefficient of nitrogen allocation, and K_{df} is the extinction coefficient for diffuse light. It is appropriate to assume that K_{df} is equal to K in Equation (10). Empirical data suggest that the ratio between K_N and K_{df} is approximately 0.4 [15]. Using the substitution $v = z_C - z$, one arrives at the following equation.

$$\begin{aligned} P(v) &= P_0 \cdot \exp\left(-0.4 \cdot K \cdot LAI \cdot \frac{v}{v+z}\right) & & \\ P(0) &= P_0 & & [\text{top of canopy}] \\ P(z_c) &= P_0 \cdot \exp(-0.4 \cdot K \cdot LAI) & & [\text{bottom of canopy}] \end{aligned} \tag{12}$$

This model means that the degree of carbon uptake will decrease as one moves from the top to the bottom of the canopy; the rapidity of the decrease will depend upon the canopy density. In the LLWR ^{14}C model interest lies not in the absolute rate of photosynthetic uptake, but rather the fractional ^{14}C uptake per unit height through the canopy. It is possible to calculate the plant uptake of ^{14}C by integrating this equation, subject to the following two assumptions. Firstly, the limits of the integral must be constrained to ensure that any fraction of the compartment that is above the plant canopy height is not considered for plant uptake of carbon. Secondly, P_0 must be defined so that $P(v)$ integrates to 1 over the range $[0, z_C]$.

Further details on this calculation, and the subsequent stages of the biosphere assessment used to calculate potential impacts to humans following a release of ^{14}C-labelled gas to the soil zone, are given in [4].

3. MODEL RESULTS

In this section the calculated plant ^{14}C concentrations resulting from a flux of 1 $Bq\,m^{-2}\,y^{-1}$ of plant available ^{14}C entering the soil are presented. These concentrations scale linearly with the flux of plant available ^{14}C. Four biosphere calculations cases are considered:

- Case 1: Assumptions as for the reference case of the 2011 ESC are used (uniform uptake of C through the canopy profile, z_d occurring at two-thirds the canopy height, 100% of any CH_4 released being oxidised to CO_2).
- Case 2: Light intensity approach used to calculate plant uptake of C [15]. Other assumptions are as reference case.
- Case 3: Assume flux to biosphere is 50% CO_2 and 50% CH_4, and that only 10% of the CH_4 released is oxidised. Other assumptions as reference case.
- Case 4: z_d position is one-sixth of the canopy height [12]. Other assumptions as reference case.

Table 1 below contains the calculated plant ^{14}C concentrations for each of these cases. Whilst the assumptions used in Cases 2 and 3 resulted in calculated ^{14}C plant concentrations which are between 5 to 45% lower than the reference case values, lowering the zero displacement layer (z_d) in Case 4 leads to a dramatic decrease of 89 to 94% in the calculated plant ^{14}C concentrations.

Table 1. Calculated ^{14}C concentration in the plants.

Plant Type	Calculated plant ^{14}C concentration (Bq kg^{-1} [C])			
	Case 1 (reference)	Case 2 (light intensity)	Case 3 (10% CH_4 oxidation)	Case 4 (z_d lower in) canopy)
Root vegetables, tubers and leafy green vegetables	$1.0\ 10^{0}$	$7.9\ 10^{-1}$	$5.5\ 10^{-1}$	$6.5\ 10^{-2}$
Garden fruit	$1.4\ 10^{0}$	$1.1\ 10^{0}$	$7.6\ 10^{-1}$	$8.9\ 10^{-2}$
Cattle and goat pasture	$5.1\ 10^{-1}$	$4.8\ 10^{-1}$	$2.8\ 10^{-1}$	$3.4\ 10^{-2}$
Sheep pasture	$5.3\ 10^{-2}$	$5.1\ 10^{-2}$	$2.9\ 10^{-2}$	$5.8\ 10^{-3}$

4. DISCUSSION

The LLWR is an active participant within the ^{14}C working group of the BIOPROTA international forum (the work of this group is presented in [5, 16]). Within this working group, a range of models used by waste management organisations have been applied to a 1 Bq m^{-2} y^{-1} ^{14}C-labelled gas release scenario, as used in the results presented above. The results presented above are therefore discussed in the context of the results of the BIOPROTA ^{14}C working group.

The model developed for the LLWR differs from all the others applied to the scenario in that it has explicit representation of two layers within the canopy atmosphere, and the plant uptake of carbon from each layer. Although the calculated ^{14}C concentration in the upper canopy layer lies within the range of values calculated by the other models applied to the scenario, the calculated ^{14}C concentration of the lower canopy atmosphere layer (that subject to diffuse mixing of air only) is more than a factor of two (and up to around three orders of magnitude) greater than the calculated ^{14}C concentrations of the single atmosphere compartments of the other models. The difference of three orders of magnitude arises in comparison with a model in which the atmosphere is considered well-mixed from the ground surface to a substantial height above the top of the plant canopy. In that model, rapid dilution occurs with the advective flow of air passing over the canopy. Thus, that model is not well-adapted to representing the situation occurring within a dense canopy, as is addressed in the model developed for the LLWR. As a result of the high calculated ^{14}C in the lower canopy atmosphere layer the calculated plant ^{14}C concentrations using the model developed for the LLWR are at least one order of magnitude greater than those calculated by the other models.

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