

# Kinetic Models for Representing the Uptake of Radionuclides in Plants

M. C. Thorne

**Abstract** Traditionally, radionuclide accumulation in plants due to uptake from soil has been estimated using empirically measured concentration ratios. However, over the last 40 years, there has been an increasing interest in representing radionuclide transport in soils and uptake by plants using a kinetic approach. Here, a brief account is given of the processes involved and of the fundamental equations used to represent the kinetics of radionuclide transport and plant uptake. In this account, the focus is on developing an appropriate representation of soil hydrology to provide water fluxes for use in the advective–dispersive transport equation. Following this account, a description is given of various models that have been used to represent radionuclide transport in soil–plant systems. These models illustrate how a wide variety of factors such as sorption, active uptake by roots, plant growth, changing hydrological conditions and volatilisation have been taken into account. In addition, a summary is given of how plant uptake of  $^{14}\text{C}$  can be estimated when the  $^{14}\text{C}$  enters the soil zone from below as either carbon dioxide or methane.

**Keywords** Radionuclides · Soil–plant model · Kinetics · Soil hydrology · Bioturbation · Volatilisation · Carbon-14

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## 1 Introduction

Conventionally in radiological impact assessment studies, the uptake of radionuclides by plants from soil is represented using concentration ratios between plant tissues and the top layer of the soil down to a depth of 0.1 or 0.2 m. This is sometimes described as an equilibrium model, but it rather represents an empirical approach in which the measured concentration in the plant part at harvest (or at sampling for pasture, as this does not have a well-defined time of harvest) is compared with the measured concentration in soil. There is no implication that an equilibrium relationship between plant and soil concentrations exists at harvest or at the time of sampling, or even that such an equilibrium would ever be attained.

The concentration ratio approach has been extremely useful in radiological impact assessment studies relating to routine releases, but is less relevant to short-term intentional or accidental releases, when the time from deposition to harvest may be an important consideration. Furthermore, it does not provide a useful conceptual framework for investigating how transient aspects of the soil–plant system, e.g. the changing water content of the soil, influence plant uptake.

Because the uptake of contaminants by plant roots is closely related to soil properties such as water content and redox potential which also influence the migration of those radionuclides in soil, it is important to treat radionuclide transport in the soil and uptake by plants in an integrated fashion. It is also convenient to treat radionuclide transport in plants following root uptake within the same conceptual framework.

Following this introduction, Sect. 2 describes the processes that need to be taken into account when modelling radionuclide transport in soil and uptake by plants, Sect. 3 then describes an early approach to the kinetic modelling of agricultural soil–plant systems and how this was taken forward in a subsequent model. Section 4 then describes some more recent models of such systems. Modelling the uptake of  $^{14}\text{C}$  from soils raises some special issues, and these are addressed in Sect. 5. Finally, Sect. 6 provides some conclusions and identifies potential directions in which this field might develop. To keep the discussion to a reasonable length, models for radionuclide transfers in forest ecosystems are not addressed in this chapter.

## 2 Processes to be Represented

For most radionuclides, transport in the soil column is by advection–dispersion in soil solution. Thus, a prerequisite for modelling radionuclide transport is an appropriate representation of the flow of soil water. Conventionally, this is addressed through the use of Richards' equation (Richards 1931). In one dimension vertically, this can be written in terms of two relationships.

$$\partial\theta/\partial t = -\partial q/\partial z \quad (1a)$$

$$q = -K(\theta)\partial\{\psi(\theta) + z\}/\partial z = -K(\theta)\{\partial\psi(\theta)/\partial z + 1\} \quad (1b)$$

where  $\theta$  (–) is the fractional water content of the soil,  $t$  (s) is time,  $q$  ( $\text{m s}^{-1}$ ) is the flow rate of water in the soil,  $z$  (m) is the vertical height in the soil column,  $K(\theta)$  ( $\text{m s}^{-1}$ ) is the unsaturated hydraulic conductivity of the soil and  $\psi(\theta)$  (m) is the pressure head.

The form of Eq. (1a, 1b) has been used to emphasise that the advective water flow rate is implicit in Richards' equation. Note that the velocity of water flow is given by  $q/\theta$ .

The hydraulic conductivity and the pressure head are both functions of the water content. Various parameterisations have been specified for the relationship between water content and the pressure head (or suction), e.g. those of Brooks and Corey (1964) and van Genuchten (1980), as discussed by Fredlund and Xing (1994) and Wheeler et al. (2007). Specifically, the van Genuchten (1980) relationship is conventionally written (Wheeler et al. 2007) as:

$$S_e = (\theta - \theta_r)/(\theta_s - \theta_r) = [1/(1 + |\alpha\psi(\theta)|^n)]^{-(1-1/n)} \quad \text{for } \psi(\theta) < 0 \quad (2)$$

where  $S_e$  (–) is the degree of saturation,  $\theta_s$  (–) is the saturated moisture content,  $\theta_r$  (–) is the residual moisture content and represents the soil moisture that cannot be removed by capillary suction, and  $\alpha$  and  $n$  are model parameters.

A generic method for obtaining unsaturated hydraulic conductivities from the degree of saturation was developed by Mualem (1976). When applied to the van Genuchten (1980) relationship, this gives (Wheeler et al. 2007)

$$K(\theta) = K_s S_e^\eta \left[ 1 - (1 - S_e^{1/m})^m \right]^2 \quad (3)$$

where  $\eta$  is a tortuosity factor obtained through calibration against observations (Schaap and Leij 2000).

In practice, for a vegetated soil, Eq. (1a) needs to be modified to allow for water uptake by plant roots (Wheeler et al. 2007). Thus,

$$\partial\theta/\partial t = -\partial q/\partial z - u_w \quad (4)$$

where  $u_w$  ( $\text{s}^{-1}$ ) is the rate of water uptake by plant roots.

When simulating radionuclide transport, it is conventional to represent sorption to soil solids by an equilibrium distribution coefficient or  $Kd$  ( $\text{m}^3 \text{kg}^{-1}$ ) value (IAEA 2010). This approach is convenient because it allows transport to be described using a single equation. However, in more comprehensive models,  $Kd$  values can be replaced by kinetic rate coefficients representing sorption and desorption, respectively. In such models, transport in soil solution is represented by one equation and transport on soil solids, e.g. by bioturbation, by a second. Using the equilibrium sorption approach, the transport equation can be written (Wheeler et al. 2007):

$$\partial[\theta C + S]/\partial t = -\partial[J_d + J_q]/\partial z - u_s - \Gamma \quad (5)$$

where  $C$  ( $\text{Bq m}^{-3}$ ) is the concentration in soil solution,  $S$  ( $\text{Bq kg}^{-1}$ ) is the concentration in soil solids,  $J_d$  ( $\text{Bq m}^{-2} \text{s}^{-1}$ ) is the dispersive flux,  $J_q$  ( $\text{Bq m}^{-2} \text{s}^{-1}$ ) is the advective flux,  $u_s$  ( $\text{Bq m}^{-3} \text{s}^{-1}$ ) is the rate of uptake by plant roots and  $\Gamma$  ( $\text{Bq m}^{-3} \text{s}^{-1}$ ) is the rate of radioactive growth or decay. Using equilibrium sorption,

$$S = \rho_b K_d C \quad (6)$$

where  $\rho_b$  ( $\text{kg m}^{-3}$ ) is the dry bulk density of the soil.

The dispersive and advective fluxes are given by

$$J_d = -\theta D_h \partial C / \partial z \quad \text{and} \quad J_q = qC \quad (7)$$

where  $D_h$  ( $\text{m}^2 \text{s}^{-1}$ ) is the sum of the soil molecular diffusion coefficient, taking into account the tortuous nature of the solute pathways, and the mechanical dispersion coefficient, which is often written as  $d_L q / \theta$ , where  $d_L$  (m) is the soil dispersivity.

Wheater et al. (2007) used the work of Epstein (1966), Nye and Tinker (1977) and Barber (1984) to argue that the active uptake of various chemicals (including radionuclides) across the root boundary can be represented by Michaelis–Menten kinetics, i.e.

$$F = F_{\max} C_b / (k_m + C_b) \quad (8)$$

where the concentration at the root boundary  $C_b$  ( $\text{mol m}^{-3}$ ) can be equated to the concentration in bulk soil solution,  $F$  ( $\text{mol s}^{-1} \text{m}^{-2}$ ) is the flux per unit area of root boundary and  $F_{\max}$  ( $\text{mol s}^{-1} \text{m}^{-2}$ ) and  $k_m$  ( $\text{mol m}^{-3}$ ) are coefficients specific to the chemical under consideration.

In practice, radionuclides are normally present in trace quantities in soils, so Eq. (8) can be linearised to give

$$F_r = \alpha C \quad (9)$$

where  $F_r$  ( $\text{Bq s}^{-1} \text{m}^{-2}$ ) is the radionuclide uptake rate per unit area of root,  $\alpha$  ( $\text{m s}^{-1}$ ) is the root absorbing power (Nye 1966) and  $C$  ( $\text{Bq m}^{-3}$ ) is the radionuclide concentration in soil solution, as defined previously.

From this, it follows that

$$u_s = 2\pi a \rho_r \alpha C \quad (10)$$

where  $a$  (m) is the mean root radius and  $\rho_r$  ( $\text{m m}^{-3}$ ) is the total root length per unit volume of soil.

The approach outlined above has some limitations. Specifically, it treats sorption as being a reversible process. This ignores the possibilities that radionuclides can be incorporated irreversibly within soil solids, may be precipitated or co-precipitated from soil solution and may be incorporated in organic matter in

soils from which they may be released only through mineralisation of the organic matter. Also, it ignores the possibility that some radionuclides, such as isotopes of selenium, may be volatilised from soils and plants (Sheppard and Sheppard 2008; see also Pérez-Sánchez et al. 2012).

Bioturbation also has to be considered in modelling the transport of radionuclides in soils. In agricultural soils, mass transport of soil solids is primarily mediated by earthworms. Shallow-burrowing earthworms are predominantly active in the top 10–25 cm and consume earth, primarily burrowing in a horizontal direction. Deep-burrowing earthworms mainly occur in the top 45 cm and primarily burrow in a vertical direction, coming to the surface to ingest foliage and other organic matter. The deepest burrowing earthworm is *Lumbricus terrestris*, which is mainly found within the top 1 m, but has been observed at depths of up to 2.5 m (Bishop 1989).

Bishop (1989) noted that the amount of earthworm activity is dependent on the type of soil, the type of land use and the time of year. The habitats likely to support the greatest densities of earthworm population are base-rich pastures and orchards where the soil is fairly well drained and loamy in texture. The lowest densities are associated with acid moorlands where the soils are shallow and peaty. Earthworm activity is found to be high in both the spring and the autumn and low during the summer and winter.

For a one-dimensional vertical soil model, the bioturbation rate needs to be based on the amount of soil redistributed vertically rather than the total amount of soil ingested and egested by earthworms. This can be estimated from the total amount of earthworm casts deposited on the surface. Bishop (1989) reports a range of 0.25–6.18 kg m<sup>-2</sup> y<sup>-1</sup> for a variety of arable and pasture farmland. Müller-Lehmans and van Dorp (1996) reported a range of 1.88–13.40 kg m<sup>-2</sup> y<sup>-1</sup> for European pasture and recommended a value of 6 kg m<sup>-2</sup> y<sup>-1</sup>, with 2 kg m<sup>-2</sup> y<sup>-1</sup> coming from deeper soil (greater than 25 cm in depth).

Recently, Klos et al. (2014) have introduced bioturbation into a radionuclide transport model in which the soil column is divided into 0.2-m-thick layers. In this model, the rate coefficients between the soil layers were augmented by adding a term  $M_{i,j}/\rho_i d_i$ , where  $M_{i,j}$  (kg m<sup>-2</sup> y<sup>-1</sup>) is the rate at which soil is moved from layer  $i$  to layer  $j$  by bioturbation (expressed on a dry weight basis),  $\rho_i$  (kg m<sup>-3</sup>) is the dry bulk density of soil in layer  $i$  and  $d_i$  (m) is the depth of layer  $i$ . In general, mass balance considerations dictate that  $M_{i,j} = M_{j,i}$  for any  $i$  and  $j$ . The values of  $M_{i,j}$  used by Klos et al. (2014) are listed in Table 1.

### 3 An Early Approach to Kinetic Modelling and Subsequent Developments to that Approach

Prior to the early 1980s, soil–plant models tended to be very simple. The soil was typically treated as a single well-mixed layer, and plant uptake was estimated by applying a plant/soil concentration ratio to the radionuclide concentration

**Table 1** Bioturbation rates used by Klos et al. (2014). Soil layer thicknesses are 0.2 m

Parameter	Value ( $\text{kg m}^{-2} \text{y}^{-1}$ )	Parameter	Value ( $\text{kg m}^{-2} \text{y}^{-1}$ )
$M_{1,2}$	0.54	$M_{2,1}$	0.54
$M_{2,3}$	0.42	$M_{3,2}$	0.42
$M_{3,4}$	0.30	$M_{4,3}$	0.30
$M_{4,5}$	0.18	$M_{5,4}$	0.18
$M_{5,6}$	0.06	$M_{6,5}$	0.06

estimated for that layer. External contamination of plants was represented by applying a single exponential loss function to the initially deposited activity, and little consideration was given to translocation of external deposits to internal plant tissues (Coughtrey and Thorne 1983). However, in 1983, Coughtrey and Thorne (1983) described new dynamic mathematical models for simulating radionuclide transport in soils, plants and domestic animals. These models (the SPADE system) took account of the need to be able to simulate a wide variety of different agricultural practices over a wide range of timescales. The soil–plant model concentrated on the topsoil. Thus, in the model, the soil was distinguished into ten layers each 0.03 m thick. In each layer, three components were identified: soil solution, organic matter and inorganic matter. Downward transfers were postulated for each of these components; that is, the model was developed only for initial deposition at the surface, and the mechanisms of transfer were not specified. Within each layer, exchange processes were postulated between soil solution and both organic and inorganic matter. Thus, soil solution acted as the medium of exchange between these other components.

In the SPADE system, plants were modelled as being composed of up to six well-mixed compartments. These comprised root, ‘root store’, stem, internal leaf, external leaf and grain or fruit. The ‘root store’ compartment was introduced so that the model could represent root and tuber crops, as well as allowing distinctions to be made between annual, biennial and perennial species. Leaves were distinguished into external and internal components to allow particulate deposition to be distinguished from incorporation.

In order to limit the number of rate coefficients that had to be specified separately, some simple rules were introduced governing the rate of transport in soil solution (constant with depth), the rates of exchange between soil solution and both organic and inorganic matter (exponential variation with depth), the rates of vertical transport of both organic and inorganic matter (exponential variation with depth) and the rate of root uptake (multi-exponential variation with depth).

The SPADE modelling system was used by the UK Ministry of Agriculture, Fisheries and Food (MAFF) and latterly by the Food Standards Agency (FSA) through to about 2005. Subsequently, it has been replaced by the PRISM model. This was initially described in a set of conference papers (Maul et al. 2005; Thorne et al. 2005; Robinson et al. 2005), and the most recent version is detailed in full in a set of technical reports (Walke et al. 2012a, b; Watson 2012).

The soil–plant model adopted in PRISM is substantially different from that in SPADE and is described in some detail below.

To represent the soil, ten soil layers are again used, so that the model has a well-defined compartmental structure. Typically, the superficial litter layer will be thin ( $\sim 0.01$  m), and this is layer 1. For farmed soils, the A horizon is typically about 0.3 m thick. To maintain sensible vertical discrimination, this is split into 3 layers of thickness 0.1 m (layers 2–4). The B horizon plus substrate goes down to about 2 m to conform to the HOST hydrological scheme that is applicable to the whole of Britain (Institute of Hydrology 1995). However, cereal roots are likely to penetrate to depths of up to 1 m and deep ploughing could go to a depth of more than 0.31 m, so it is necessary maintain more discrimination in the top of the zone. Layers 5–8 have a thickness of 0.2 m and layers 9 and 10 thicknesses of 0.45 m. This gives an overall soil profile depth of 2.01 m.

For the purpose of assigning properties, each layer is considered to be of one of three textures: sand, clay or loam.

Plants are distinguished into seven components, as shown in Fig. 1. Transfer rates between the different plant components are specified directly in the model based on empirical observations, but, in broad terms, a distinction is made between elements that are actively transported in the phloem and those that are not (Walke et al. 2012b).

The water content of each of the vertical compartments in the soil is computed by mass balance, taking account of precipitation input ( $P$ ), evapotranspirative losses ( $E_i$ ), flows between layers ( $I_{i,i+1}$  and  $I_{i+1,i}$ , with the special case to groundwater ( $G$ ) at the base of the model) and sub-horizontal drainage ( $H_i$ ).

A feature of the PRISM model is that plant growth is explicitly represented. The plant types included in the model are mixed pasture, leguminous pasture, non-leguminous fodder crops, leguminous fodder crops, cereals (including winter-sown and spring-sown options), non-leguminous green vegetables, leguminous green vegetables, root vegetables, tubers, tree fruit, shrub fruit and herbaceous fruit. Growth functions are defined for individual plant components (foliage, stem, fruit/grain and roots) and require the final aboveground biomass ( $W$ ), the time when growth starts for each component ( $t_{\text{start}}$ ), the time when growth ceases for each component ( $t_{\text{end}}$ ) and a growth power ( $N$ ).

The general form of the growth model for any plant component is

$$w(t) = 0 \quad t \leq t_{\text{start}} \quad (11a)$$

$$= W[(t - t_{\text{start}})/(t_{\text{end}} - t_{\text{start}})]^N \quad t_{\text{start}} \leq t < t_{\text{end}} \quad (11b)$$

$$= W \quad t \geq t_{\text{end}} \quad (11c)$$

where  $w(t)$  is the mass of the component at time  $t$ .

The value of  $N$  is typically 2.2.

The PRISM root profiles are determined based on root-shape modifiers, which are defined as the ratio of root density in a particular soil layer in field conditions to

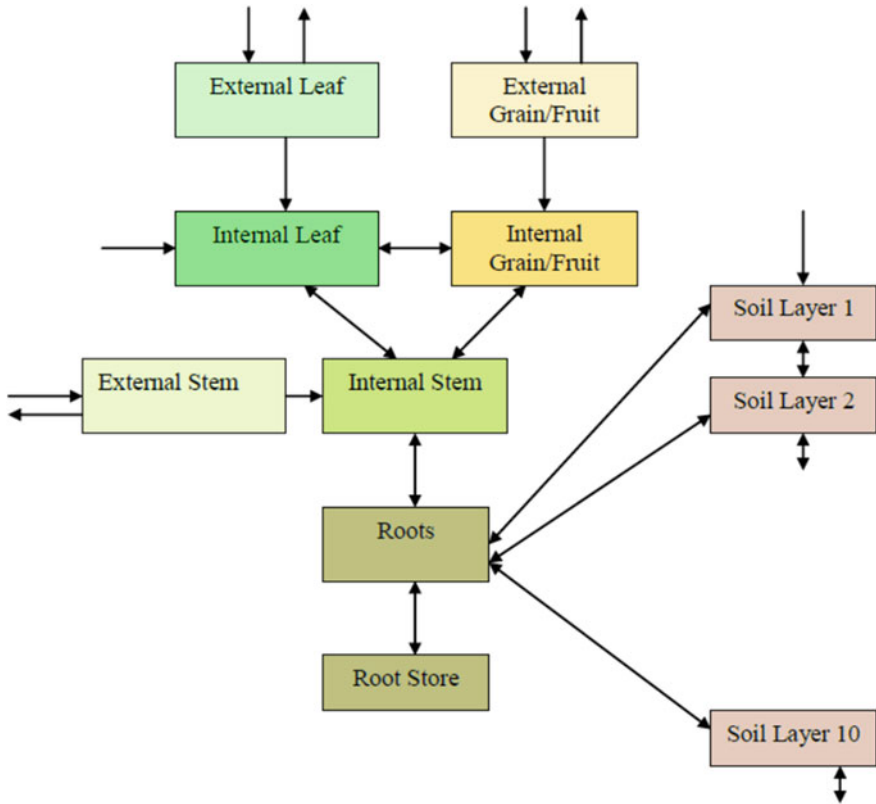


Fig. 1 Structure of the PRISM soil–plant model. (From Walke et al. 2012b)

that in pot experiments,  $s$ . Root growth is also dependent on a root profile development power,  $v$ . For a mature crop, reference values of  $s$  of 1 are used for soil layers 1–3, as a well-developed root system in the field should approximate closely to a well-developed root system in a pot, providing that the pot is not highly restrictive of the root system. Values of  $s$  for the deeper layers are defined by reference to observed root profiles. These are primarily taken from Bishop and Beetham (1989). The equation representing root growth in a soil layer is

$$s(t) = S[w_r(t)/W_r]^v \tag{12}$$

where  $w_r(t)$  is calculated as in Eqs. (11a, 11b, 11c) and  $S$  is the value of  $s$  that applies when the plant is mature. The development of  $s(t)$  for winter-grown cereals is illustrated in Fig. 2.

Alternatively, an exponential root-shape model may be used. Such a model was developed by Gerwitz and Page (1974) and was applied by Wheeler et al. (2007) to



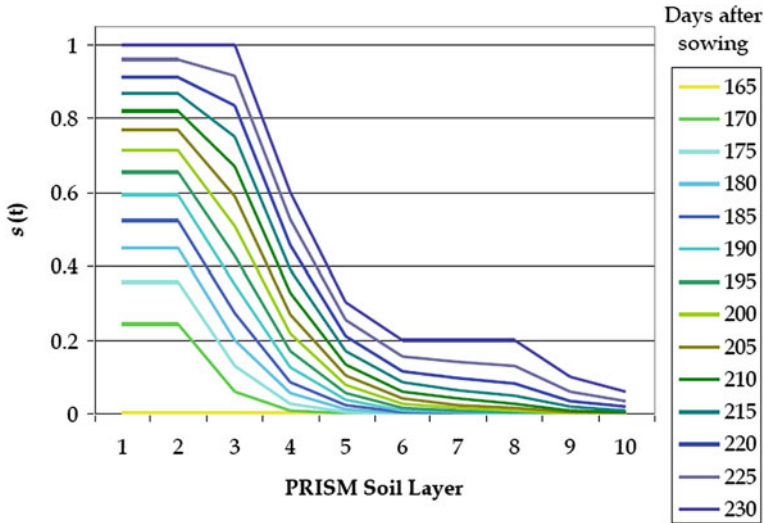


Fig. 2 Development of root-shape modifiers for winter cereals. (From Walke et al. 2012b)

a series of root density measurements of wheat taken by Burne et al. (1994). This model has the form

$$\rho_r(z, t) = \rho_{r0}(t)\exp[z/z_r(t)] \tag{13}$$

where  $\rho_{r0}(t)$  is the root density at the soil surface at time  $t$ ,  $z$  is the depth from the surface and  $z_r(t)$  is the time-dependent root attenuation coefficient.

An important innovation in the PRISM model was that it used the degree of sorption of radionuclides to soil solids to adjust empirically observed plant–soil concentration ratios to make them of wider applicability. The argument is based on the consideration that with equilibrium, reversible sorption, a contaminant is partitioned between soil solution and soil solids. The uptake affinity,  $\delta$ , is defined relative to soil solution containing unit concentration of the radionuclide. Thus, the plant/soil concentration ratio,  $CR$ , as conventionally defined is given by'

$$CR = \delta/(\theta + \rho_b Kd) \tag{14}$$

hence, Eq. (14) can be inverted to give

$$\delta = CR(\theta + \rho_b Kd) \tag{15}$$

Values of  $\delta$  are found to be much more consistent for individual radionuclides than are  $CR$  values. Furthermore, they allow the effects of time-dependent changes in soil moisture content,  $\theta$ , to be explicitly taken into account.

## 4 More Recent Modelling Approaches

Based on some of the developments included in the PRISM model and other considerations specific to the behaviour of selenium in soils, Pérez-Sánchez et al. (2012) developed a kinetic model to represent the transport of  $^{79}\text{Se}$  in soils and its uptake by plants.

As discussed by Pérez-Sánchez et al. (2012), within soils, selenium can be thought of as most resembling sulphur in its chemical behaviour (Wheater et al. 2007). Selenium exists as anionic complexes in groundwater and in four oxidation states: selenate(+6), selenite(+4), elemental Se(0) and selenide(-2) (Fio et al. 1991). The speciation of selenium is dependent upon a number of factors, such as pH, the soil redox potential (Eh), the mineralogical and chemical composition of the soil, microbial interactions and the nature of adsorbing surfaces (Neal 1995). However, redox potential exerts the most influence over selenium behaviour in soil (Moore 2004). Thus, in very oxidising conditions, selenate ( $\text{SeO}_4^{2-}$ ) will tend to dominate selenium speciation (Fio et al. 1991; Neal 1995). As the redox potential falls under more saturated, reducing conditions, selenate will be successively reduced to selenite ( $\text{SeO}_3^{2-}$ ), elemental selenium and selenide. This has important implications for the bioavailability of selenium (Moore 2004); the more reducing the soil conditions, the less soluble and mobile selenium becomes (Neal 1995).

Additionally, there is extensive evidence that selenium is volatilised from both soils and plants (Pérez-Sánchez et al. 2012; Sheppard and Sheppard 2008). Furthermore, selenium incorporated in plants can be transferred to the organic matter 'pool' in soil whence it will be returned to soil solution by mineralisation processes.

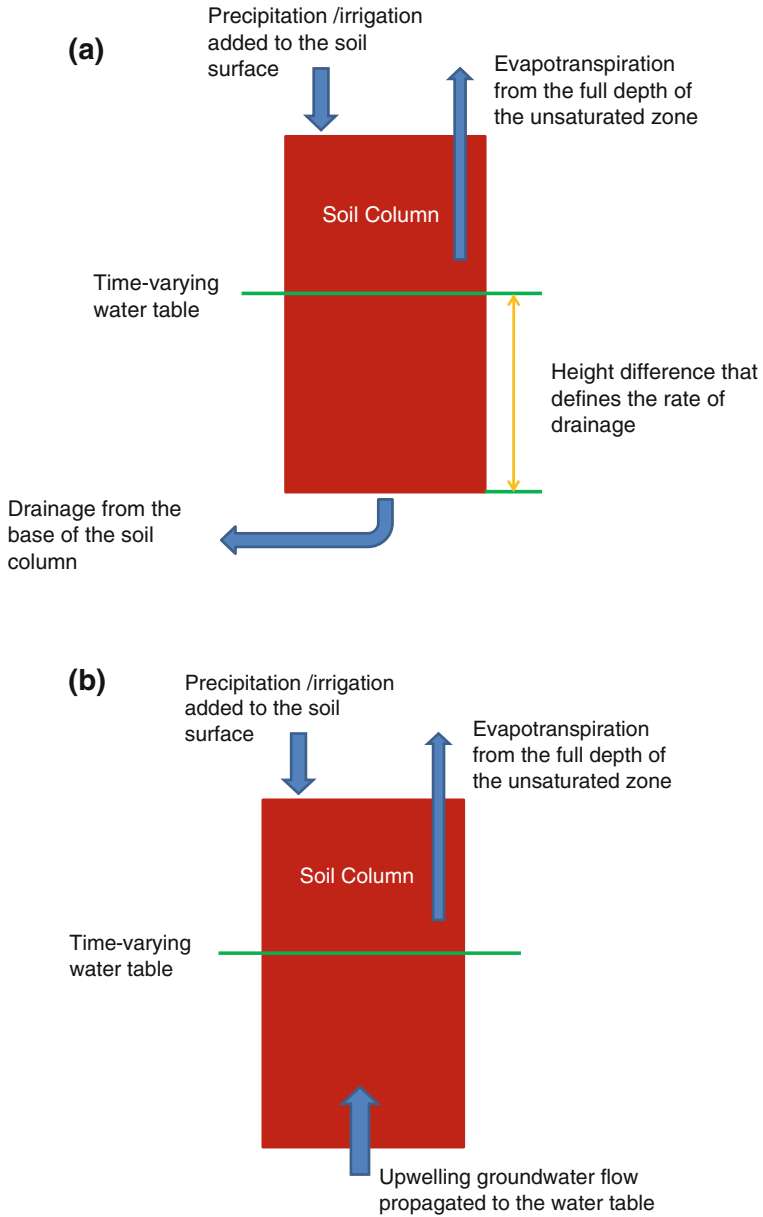
Based on the above considerations, Pérez-Sánchez et al. (2012) included in their model changes in soil hydrology and their effects on redox conditions, volatilisation from soils and plants, and the incorporation of selenium in organic matter in soil and its subsequent release. Of particular interest is the simplified approach to representing soil hydrology that they developed.

In the adopted approach, a 1D soil column is used. This soil column is considered either to drain from its base or to receive an upward groundwater flux from below. The former is appropriate to well-drained agricultural soils, whereas the latter is appropriate to discharge zones (e.g. riparian areas). These two situations are illustrated in Fig. 3.

The fundamental equation used to represent the situation shown in Fig. 3a is

$$dh/dt = [R_i - ah^b - f_i PE_i] / \Delta\theta \quad (16)$$

where  $h$  (m) is the height of the water table at time  $t$ ;  $R_i$  ( $\text{m y}^{-1}$ ) is the average precipitation/irrigation rate in month  $i$ ;  $PE_i$  ( $\text{m y}^{-1}$ ) is the potential evapotranspiration rate for month  $i$ ;  $a$  and  $b$  are calibration constants;  $f_i$  is the fraction of the potential evapotranspiration ( $PE$ ) that constitutes actual evapotranspiration ( $AE$ ); and  $\Delta\theta$  is the difference in water content between soil that is fully saturated and soil that contains only some minimal water content.



**Fig. 3** Conceptual models for the hydrology of a freely draining soil column (a) and a column underlain by upwelling groundwater (b)

In the following, the month index  $i$  is generally suppressed for conciseness, but should be understood to apply throughout.

Additionally, a variable  $h_2$  is defined and  $\theta$  is allowed to vary from its value at the height of the water table ( $\theta_1$ ) to its value at height  $h_2$  ( $\theta_2$ ). Linear interpolation is used over this range. Thus,

$$\theta = \theta_2 \quad z \geq h_2 \quad (17a)$$

$$\theta = \theta_1 + (z - h)(\theta_2 - \theta_1)/(h_2 - h) \quad h < z < h_2 \quad (17b)$$

$$\theta = \theta_1 \quad z \leq h \quad (17c)$$

It is readily shown that to increase the height of the water table by unit amount in unit area, it is necessary to add a volume of water  $\theta_1 - \theta_2$ , corresponding to  $\Delta\theta$ . Thus,

$$dh/dt = [R - ah^b - fPE]/(\theta_1 - \theta_2) \quad (18)$$

Losses through drainage are represented through the term  $-ah^b$  with the rate of loss being a monotonically increasing function of the height difference between the water table and the base of the soil column.

However, where groundwater upwelling dominates, Eq. (18) is replaced by

$$dh/dt = [R + F - fPE]/\Delta\theta \quad (19)$$

where  $F$  is the upwelling flux of groundwater ( $\text{m y}^{-1}$ ).

This approach neglects any losses by interflow downslope within the soil. This is cautious, since it maximises the upward transport of contaminants. However, if the water table reaches the surface, any excess water is taken to be expelled from the soil column and lost as overland flow. This excess water comprises both upwelling groundwater and precipitation that has never penetrated the soil column.

In the model, changes in the height of the water table are calculated on a monthly basis, with  $R$  and  $PE$  also specified on a monthly basis and  $F$  (as required) specified as a long-term constant value. Thus, in the groundwater-upwelling variant,

$$\Delta h = \Delta t[R + F - fPE]/\Delta\theta \quad (20)$$

In order to account for surface runoff when  $h = H$  (i.e. the soil is saturated to the surface at height  $H$ ),

$$h_n = \min(h_o + \Delta h, H) \quad (21)$$

where  $h_n$  is the new value of  $h$  and  $h_o$  is the old value of  $h$ .

The rate of surface runoff,  $S$ , is given by

$$S = \max(0, \{h_o + \Delta h - H\} \Delta \theta) \quad (22)$$

Having defined the location of the water table using the above approach, water fluxes between the various soil layers are computed differently for the zone beneath the water table and the zone above the water table. Above the water table, the water flux at height  $z$ ,  $F(z)$ , is computed in the same way for both the draining and recharging cases, i.e.

$$F(z) = R - fPE + fPE(z - h)/(H - h) \quad H \geq z \geq h \quad (23)$$

However, below the water table, there may be a time-dependent drainage flux,  $D$ , or a constant upwelling flux,  $F$ . Thus, for cases 3a and 3b, the following alternative expressions apply:

$$F(z) = D = ah^b \quad h > z \geq 0 \quad (24a)$$

$$F(z) = -F \quad h > z \geq 0 \quad (24b)$$

Note that the convention used is that positive fluxes are directed downwards.

As water contents and water fluxes are defined in and between all the layers of the soil column, radionuclide transport can be represented using the advective–dispersive transport approach outlined in Sect. 2. In practice, in the model, the dispersive component is neglected and a purely advective approach is adopted. Sorption is taken into account using an equilibrium approach; that is, the effective volume of each compartment includes the effects of sorption. The model is normalised to a reference soil area,  $A$ , of 1 m<sup>2</sup>. Thus, the effective volume of each soil layer  $i$ ,  $V_i$  (m<sup>3</sup>), is given by

$$V_i = Ad_i(\theta_i + \rho Kd_i) \quad (25)$$

where  $d_i$  (m) is the user-specified depth of the soil layer;  $\theta_i$  (–) is the average water content of that soil layer obtained from the hydrological model;  $\rho$  (kg m<sup>–3</sup>) is the dry bulk density of the soil; and  $Kd_i$  (m<sup>3</sup> kg<sup>–1</sup>) is the distribution coefficient for that soil layer.

Above the capillary fringe, oxic conditions prevail and one value of the distribution coefficient ( $Kd_2$ ) applies. Below the water table, anoxic conditions prevail and a second distribution coefficient ( $Kd_1$ ) prevails. Between, a linear interpolation is assumed, i.e. at height  $z$ , where  $h_2 > z > h$ :

$$Kd(z) = Kd_2 + (Kd_1 - Kd_2)(h_2 - z)/(h_2 - h) \quad (26)$$

From the above, it follows that

$$\lambda_{ij} = AF_{ij}/V_i \quad (27)$$

where  $\lambda_{ij}$  ( $y^{-1}$ ) is the rate coefficient for transfer from layer  $i$  to layer  $j$  (defining layer 0 as a sink for surface discharge and layer 11 as a sink for drainage through the base of the soil column).

By substitution from Eq. (25),

$$\lambda_{ij} = F_{ij}/d_i(\theta_i + \rho Kd_i) \quad (28)$$

Volatilisation from the soil is taken into account using a simple parameterisation, in which the volatilisation rate is dependent only upon the soil moisture content. It is argued that there are two considerations that will determine the evolution of volatile selenium from soil. These are the location of production and the ease of escape to the soil surface. Both of these will be related to the water content of the soil. The production of volatile forms is likely to be microbially mediated. As the most likely gases to be produced are methylated forms, production is likely to be concentrated at or below the water table, since methylation is unlikely to be significant in aerobic conditions (though minor anaerobic pockets may occur within the generally aerobic soil zone and volatilisation has been reported from aerobic soils). However, at depths significantly below the water table, selenium is likely to become dissolved and may be reutilised rather than released. In view of these considerations, a generalised loss rate model is specified in which the loss rate takes a constant (low) value for highly unsaturated soils, increases to a maximum for soils close to saturation and then remains constant or decreases to a constant value below the water table.

The available evidence suggests that volatilisation of selenium from plants occurs as a consequence of root uptake of selenium in solution and the subsequent production of volatile compounds within the plant (Limer and Thorne 2010). Therefore, the primary consideration is to develop a root uptake model for selenium in solution, since volatilisation can be represented as a loss process from the plant in competition with other loss processes such as cropping. The root uptake model developed by Pérez-Sánchez et al. (2012) is similar to that described above for PRISM and is not described further herein.

Cropping of plants is represented by a rate coefficient determined by the ratio of the rate of biomass removal and the standing biomass at any time.

In principle, a complex model for organic degradation and mineralisation reactions could have been included. However, Pérez-Sánchez et al. (2012) argued that because of uncertainties in how selenium mineralisation is related to carbon mineralisation, a complex approach is not justified, so a single rate constant for such degradation was defined. As the organic matter is likely to be present mainly in the upper layer of the soil system, this activity is returned to the uppermost soil layer.

Subsequently, Pérez-Sánchez and Thorne (2014) have adapted the selenium model described above to make it applicable to radionuclides in the  $^{238}\text{U}$  decay chain. This enhancement now also includes a representation of bioturbation (Pérez-Sánchez and Thorne, *Journal of Radiological Protection*, in press) following that adopted by Klos et al. (2014).

The above discussion has emphasised vertical 1D models of radionuclide transport in soils and uptake by plants. However, 2D and 3D representations are also used. In particular, 3D, physically based, surface-water catchment models can be used to represent contaminant transport both in solution and as a consequence of sediment transport (Towler et al. 2011). Such models have been extensively used in the development of safety cases for the geological disposal of radioactive wastes (Bosson et al. 2010, 2012a, b). These models are not described here, as the principles for representing contaminant transport in the soil zone and uptake by plants are the same as applied in 1D.

## 5 Transport of Carbon-14 in Soils and Uptake by Plants

When  $^{14}\text{C}$  is released to atmosphere, uptake by plants is relatively readily modelled. In general terms, the specific activity of carbon dioxide in the atmosphere at ground level is calculated and carbon with this specific activity is then used as the source material for photosynthesis and biomass production. As most plant carbon originates from foliar uptake from the atmosphere and subsequent photosynthesis, this approach is a good approximation to what physically occurs. TOCATT (Le Dizès 2005; Le Dizès et al. 2012; Aulagnier et al. 2012, 2013) is a good example of a state-of-the-art model based on these principles.

However, when  $^{14}\text{C}$  is released into the soil from below, e.g. from a near-surface facility for the disposal of radioactive wastes, different considerations apply. The  $^{14}\text{C}$  is transported through the soil atmosphere by both diffusion and advection, the latter being controlled by variations in atmospheric pressure. In addition, the  $^{14}\text{C}$  dissolves in soil solution and becomes available for root uptake. Although root uptake is generally a minor source of carbon for plants, typically contributing 1–2 % of plant carbon (BIOPROTA 2014), it can be a major source of  $^{14}\text{C}$ , if the soil atmosphere is much enriched in  $^{14}\text{C}$  relative to the aboveground atmosphere.

However, most of the  $^{14}\text{C}$  that enters the soil system from below is released into the atmosphere above the soil. There, it may be temporarily retained in the plant canopy and be available for uptake by photosynthesis. As with uptake from soil, the significance of the sub-canopy uptake of  $^{14}\text{C}$  can be enhanced by the higher specific activity of  $^{14}\text{C}$  in the sub-canopy atmosphere than the above-canopy atmosphere.

Over the last few years, several models for transport of  $^{14}\text{C}$  through soils and uptake by plants have been developed and compared (Mobbs et al. 2013). The most comprehensive model that has been developed is that produced for use in the

post-closure radiological safety assessment of the UK Low Level Waste Repository (LLWR) (see Limer et al. 2011). More recently, new field studies have been reported on the transport of  $^{14}\text{C}$ -labelled methane through soils in field conditions, its conversion to  $^{14}\text{C}$ -labelled carbon dioxide and its uptake by plants. This work is underpinning the ongoing development of detailed, process-based and simplified, assessment-level modelling of the processes involved (Appendix B to BIOPROTA 2014).

## 6 Conclusions

Over the last 40 years, kinetic models for radionuclide transport in soils and uptake by plants have developed considerably. Nevertheless, for many assessment studies, reliance is still placed upon the use of empirically determined distribution coefficients and plant–soil concentration ratios. Compilations of recommended values and distributions for these factors continue to be produced (IAEA 2010), and experimental studies typically continue to be directed to their determination. It is recognised that experimental work to underpin comprehensive, process-based models is much more complex and resource intensive than the measurement of distribution coefficients and concentration ratios (Wheater et al. 2007). However, it repays the investment in terms of greater explanatory power, applicability in a wider range of circumstances and by identifying issues, such as the effects of time-dependent hydrology, that cannot even be fully articulated in a simpler modelling framework.

The earliest kinetic models, like SPADE, required the developer and/or user to estimate rate coefficients and then input them into the model. More recent developments compute the rate coefficients, which may be time dependent, from various sources of information. This information includes the empirical estimates of distribution coefficients and plant–soil concentration ratios that were used in earlier models. However, it also includes other information that could not previously readily be used, e.g. root uptake rates from studies in nutrient solution or kinetic studies of contaminant uptake on solids from solution.

The new models emphasise the importance of modelling changes in soil hydrology and soil chemistry. Currently, the models represent soil chemistry in a highly simplified way, using soil moisture content as a surrogate for redox conditions. It is anticipated that the next generation of models will include explicit coupling of hydrological, hydrogeological and hydrogeochemical processes in the soil zone, so that a much wider range of biogeochemical processes can be represented. Such an approach, in which 3D hydrological modelling is coupled to geochemical modelling, is already being explored in the sub-surface (Piqué et al. 2013), but its extension to the soil zone will require more detailed consideration of biological processes in a highly dynamic environment than is necessary at greater depths and will require that physiologically based models of plants are adopted, as in the COUP model (Jansson and Karlberg 2001). Also, models used at the present



time typically represent a 1D soil column. Within this framework, simplifications and ad hoc approximations have to be made to ensure that a coherent hydrological situation is represented. It is anticipated that, in future, radionuclide transport in soils and uptake by plants will be more often simulated in 2D (hillslope) or 3D (surface-water catchment) contexts, by including appropriate soil–plant modules within the sorts of hydrological and hydrogeological catchment-scale models that already exist (Towler et al. 2011).

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