



Modelling the behaviour of uranium-series radionuclides in soils and plants taking into account seasonal variations in soil hydrology



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ABSTRACT

In a previous paper, a mathematical model for the behaviour of ⁷⁹Se in soils and plants was described. Subsequently, a review has been published relating to the behaviour of ²³⁸U-series radionuclides in soils and plants. Here, we bring together those two strands of work to describe a new mathematical model of the behaviour of ²³⁸U-series radionuclides entering soils in solution and their uptake by plants. Initial studies with the model that are reported here demonstrate that it is a powerful tool for exploring the behaviour of this decay chain or subcomponents of it in soil-plant systems under different hydrological regimes. In particular, it permits studies of the degree to which secular equilibrium assumptions are appropriate when modelling this decay chain. Further studies will be undertaken and reported separately examining sensitivities of model results to input parameter values and also applying the model to sites contaminated with ²³⁸U-series radionuclides.

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1. Introduction

In a previous paper (Pérez-Sánchez et al., 2012), a mathematical model for the behaviour of ⁷⁹Se in soils and plants was described. Subsequently, a review has been published relating to the behaviour of ²³⁸U-series radionuclides in soils and plants (Mitchell et al., 2013). Here, we bring together those two strands of work to describe a new mathematical model of the behaviour of ²³⁸U-series radionuclides entering soils in solution and their uptake by plants. It is emphasized that the model is not intended to apply to naturally occurring ²³⁸U series radionuclides present incorporated into the soil structure, e.g. in specific mineralogical phases. Illustrative calculations undertaken using this new model are also described. It is considered that this model will find application both in safety assessments relating to the geological disposal of radioactive wastes and in programmes relating to the clean-up of sites contaminated with Naturally Occurring Radioactive Materials (NORM). Currently, the model is being applied in studies relating to the near-surface disposal of low-level radioactive wastes, taking into account both the transport of uranium-series radionuclides in groundwater and potential human intrusion resulting in the contamination of surface soils.

Typically in post-closure assessment studies the soil zone is modelled as either one or two well-mixed compartments, with transfers between and out of them governed by first-order rate coefficients and no consideration given to seasonal or inter-annual changes in hydrological or geochemical conditions (e.g. Thorne, 2007). Plant uptake in these models is represented using an equilibrium concentration ratio approach (IAEA, 2010). About 30 years ago, a new generation of multi-compartment models was developed to more adequately address the transport of radionuclides in the soil column (Thorne and Coughtrey, 1983). However, these models remained multi-compartmental in nature and did not explicitly represent the processes taking place (Thorne, 2012). More recently, physically based models based on the use of the advection–dispersion equation for transfers in soils and Michaelis–Menten kinetics for uptake by plant roots have been developed (Wheater et al., 2007). These models are generally used in a research context for short-term simulations and typically include an explicit solution of Richards (1931) equation to represent the hydrology of the unsaturated zone. Although sorption to the soil can be represented kinetically, limitations in the available data mean that it continues to be usual to represent the partitioning of radionuclides between soil solution and soil solids using an equilibrium approach. For trace quantities in solution, the degree of sorption can be taken to be independent of the concentration in solution and a distribution coefficient, or K_d , approach can be adopted (IAEA, 2010). Also, although it has been found (e.g. Epstein,

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1966; Nye and Tinker, 1977; Barber, 1984) from experimental studies of excised roots that the active uptake of various chemicals across the root boundary can be represented by Michaelis–Menten kinetics, at low concentrations in solution, the Michaelis–Menten expression reduces to a linear form, where the flux of a radionuclide across the root surface, F ($\text{mol m}^{-2} \text{s}^{-1}$), can be calculated as αC , where α (m s^{-1}) is referred to as the root sorption coefficient or ‘root absorbing power’ (Nye, 1966; Wheeler et al., 2007) and C (mol m^{-3}) is the concentration in soil solution. From this it follows that the rate of plant uptake per unit volume of soil, f ($\text{mol m}^{-3} \text{s}^{-1}$) is given by:

$$f = 2\pi a \rho_r \alpha C \quad (1)$$

where a (m) is the root radius and ρ_r (m^{-2}) is the length of roots per unit volume (Wheeler et al., 2007).

In the model described in Section 2, sorption in soils is characterized using distribution coefficients and plant uptake is characterized using the linear version of Michaelis–Menten kinetics appropriate at low solution concentrations. The main simplification relative to a full 1D research model, such as that described by Wheeler et al. (2007), is that the Richards equation approach to soil hydrology is replaced by a simpler water-balance approach with a prescribed depth of capillary fringe.

In applying the model to radionuclides in the ^{238}U series, specific consideration must be given to the geochemistry and biogeochemistry of the radionuclides in that series. In the context of transport in soils and uptake by plants, these matters are addressed in detail by Mitchell et al. (2013).

In summary, uranium and thorium are members of the actinide series of elements. Both can occur in nature in the tetravalent oxidation state, U(IV), and their ions have similar radii. Consequently, they can substitute extensively for one another, explaining their geochemical coherence under reducing conditions. However, under oxidizing conditions, uranium forms the uranyl ion, i.e. U(VI) rather than U(IV). This means that uranium is often mobile in oxidizing conditions and separates from thorium (Chapter 10 of Faure and Mensing, 2005). Radium is an alkaline earth and these elements are only found in the environment in compounds as the $2+$ ion. These compounds tend to be insoluble and thus alkaline earths are often found in insoluble deposits. Here it is relevant to note that, in environmental contexts, radium often co-precipitates with barium as a mixed sulphate phase (Mitchell et al., 2013). In the model described in Section 2, it is assumed that barium and/or sulphate concentrations in soils are sufficiently low that co-precipitation of radium can be neglected. Radon (in this case ^{222}Rn) is a noble gas. As ^{222}Rn has a radioactive half life of 3.8 days it can be transported significant distances in soil or from soil to the overlying atmosphere (primarily in the gas phase of the unsaturated zone) before it decays. However, if ^{222}Rn remains in the soil, it decays via a sequence of short-lived progeny to ^{210}Pb , with a half life of 22.2 years. In turn, the ^{210}Pb decays to ^{210}Po with a half life of 138.4 days. In terms of geochemistry, lead is a metal and a member of group IVa of the periodic table, but in many properties it resembles the alkaline earths more than other members of this group, except that its halides, hydroxides and phosphates are insoluble (Gerber et al., 1980). Polonium is a reactive, silver-grey semi-metal. It dissolves in dilute acids to form simple salts, such as the chloride, and can be present as either Po(II) or Po(IV) ions. It tends to be mobile in environmental media (Mitchell et al., 2013).

Uranium and thorium are not required for plant nutrition and their uptake by plant roots is generally very limited. Although radium is not an essential trace element for plants, it is a member of the alkaline earths, a group that contains calcium and magnesium, both of which are important for plant nutrition. The relationship of

radium uptake by plants to that of other alkaline metals differs between crops and under various experimental conditions. However, there is a reasonable consensus that the behaviour of radium in the soil–plant system is closer to that of barium than to the other alkaline earth elements (Mitchell et al., 2013). Overall, radium is moderately bioexcluded by plant roots, but not to the same degree as is observed for uranium and thorium (Mitchell et al., 2013). Lead and polonium are not required for plant nutrition and are strongly excluded from root uptake (Mitchell et al., 2013).

2. Mathematical structure of the model

The mathematical structure of the model is essentially identical to that described previously for ^{79}Se . That model was implemented in the AMBER 5.4 simulation system (Quintessa, 2012) that provides a simple method for encoding decay chains of any length. This meant that extending from single-member decay for ^{79}Se to the full decay chain for ^{238}U did not require any structural changes to the model.

2.1. Hydrology

In terms of hydrology, the fundamental equation used in the model to represent water flow in a 1D soil column is:

$$dh/dt = [R_i - ah^b - s_i PE_i] / \Delta\theta \quad (2)$$

where h (m) is the height of the water table at time t ; R_i (m y^{-1}) is the average precipitation/irrigation rate in month i ; a and b are calibration constants; PE_i (m y^{-1}) is the potential evapotranspiration rate for month i ; and s_i is the fraction of potential evapotranspiration that constitutes actual evapotranspiration in month i . $\Delta\theta$ is the difference in fractional water content between soil that is fully saturated and soil that contains only some minimal water content. Losses through drainage are represented through the term $-ah^b$. When groundwater upwelling dominates, Equation (2) is replaced by:

$$dh/dt = [R_i + F - s_i PE_i] / \Delta\theta \quad (3)$$

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

The model includes a provision for removing surface runoff when the water table height reaches the soil surface located at height H m. Details are given in Pérez-Sánchez et al. (2012). As the water table height did not reach the surface in the simulations undertaken in this study, this aspect of the model is not described herein.

Additionally, a variable h_2 is defined and θ is allowed to vary from its value at the height of the water table (θ_1) to its value at height h_2 (θ_2). Linear interpolation is used over this range. It should be noted that $\Delta\theta$ in Equations (2) and (3) corresponds to $\theta_1 - \theta_2$. It is recognized that this is a simplification of the profile that would be developed in the capillary fringe in a full Richards' equation approach. However, it is argued that this simplification is appropriate because it is seldom possible to achieve an accurate parameterization of the non-linear equations in matric potential, unsaturated conductivity and water content that govern flow in this domain.

Water fluxes are computed differently for the zone below the water table and the zone above the water table. Above the water table, the flux in month i at height z , $F_i(z)$ (m y^{-1}), is computed in the same way for both the draining and recharge cases:

$$F_i(z) = R_i - s_i PE_i + s_i PE_i (z-h) / (H-h) \quad H \geq z \geq h \quad (4)$$

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