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Coupling the advection-dispersion equation with fully kinetic reversible/irreversible sorption terms to model radiocesium soil profiles in Fukushima Prefecture

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ABSTRACT

Radiocesium is an important environmental contaminant in fallout from nuclear reactor accidents and atomic weapons testing. A modified Diffusion-Sorption-Fixation (mDSF) model, based on the advection-dispersion equation, is proposed to describe the vertical migration of radiocesium in soils following fallout. The model introduces kinetics for the reversible binding of radiocesium. We test the model by comparing its results to depth profiles measured in Fukushima Prefecture, Japan, since 2011. The results from the mDSF model are a better fit to the measurement data (as quantified by R^2) than results from a simple diffusion model and the original DSF model. The introduction of reversible sorption kinetics means that the exponential-shape depth distribution can be reproduced immediately following fallout. The initial relaxation mass depth of the distribution is determined by the diffusion length, which depends on the distribution coefficient, sorption rate and dispersion coefficient. The mDSF model captures the long tails of the radiocesium distribution at large depths, which are caused by different rates for kinetic sorption and desorption. The mDSF model indicates that depth distributions displaying a peak in activity below the surface are possible for soils with high organic matter content at the surface. The mDSF equations thus offers a physical basis for various types of radiocesium depth profiles observed in contaminated environments.

1. Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant contaminated land across Fukushima Prefecture (Evrard et al., 2015). The main nuclide of concern over the long-term from the perspectives of radiation protection and radioecology is the radiocesium isotope ^{137}Cs, which is a characteristic component of nuclear weapons-testing and reactor accident fallout. Soil consists of organic and inorganic matter that have various microscopic binding sites for radiocesium. It is well known that frayed edge sites of micaceous minerals strongly bind radiocesium (e.g., Okumura et al., 2013; Fuller et al., 2015). On the other hand, Mukai et al. (2014) reported that radiocesium sorption was not limited to frayed edge sites but occurs uniformly on weathered biotite. Murota et al. (2016) reported evidence for several radiocesium binding sites on Fukushima soil particles.

Despite its affinity for binding to soil, radiocesium gradually migrates deeper into the ground over time (Gale et al., 1964). This results in a faster rate of reduction of air dose rates (groundshine) than would otherwise be expected by radioactive decay (International Atomic Energy Agency, 2009; Saito and Onda, 2015; Mikami et al., 2015; Malins et al., 2016). The depth profile and fixation of radiocesium in soil affects its uptake by vegetation (Beresford et al., 1992; Krouglov et al., 1996) and its redistribution by soil erosion and sediment transport (He and Walling, 1997). It is important therefore to understand the physiochemical processes altering radiocesium depth distributions in soil to predict how they

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will evolve over time and to evaluate countermeasures against the contamination.

Measured profiles of the radiocesium activity with depth in soil are often fitted with empirical functions such as the exponential for the years following fallout (Beck, 1966; International Commission on Radiation Units and Measurements, 1994). This facilitates the extraction of parameters characterizing the penetration of the contamination into the soil, such as relaxation masses per unit area. Yesin and Cakir (1989), Clouvas et al. (2013), Matsuda et al. (2015), Takahashi et al. (2015) and Malins et al. (2016) are examples of such studies after the Chernobyl and Fukushima nuclear accidents.

Compared to the single exponential, Antonopoulos-Domis et al. (1995) showed that depth profiles of Chernobyl radiocesium in Northern Greece were better fitted as the sum of two exponentials, corroborating experiments by Ohnuki and Tanaka (1989). Some radiocesium depth profiles from Fukushima Prefecture also display this two exponential characteristic, where there is a steep decrease in the radiocesium activity below the surface of the ground, followed by a long tail in the distribution deeper in the soil (e.g., Matsuda et al., 2015; Takahashi et al., 2015). A third class of empirical functions, such as the Lorentz (Hillmann et al., 1996) and the hyperbolic secant (Matsuda et al., 2015), are fitted to depth profiles displaying a peak in radiocesium activity below the surface. The drawback of empirical models for the depth distribution is that they do not offer a physical basis for the formulae they employ. A similar criticism has been noted against compartment models for the radiocesium depth profile (Kirchner, 1998).

Various physiochemical models have been proposed for the evolution of radiocesium distributions within soil. Typically the models describe how the distribution along one direction (the depth direction) changes over time. The radiocesium inventory reduces by radioactive decay. Bossew and Kirchner (2004) summarized the various assumptions and limitations inherent in these models, such as the assumption of constant physical (e.g. soil density) and chemical (e.g. distribution coefficients) parameters over both soil depth and time. The main differences between the models lie in the number and types of sorption sites for radiocesium, the use of explicit kinetic terms to model sorption processes, and the inclusion of advective transport. Table 1 summarizes the key physiochemical models from the literature and the differences between them.

In the simplest case, the radiocesium migration is modelled as a diffusion process of the fraction dissolved within soil pore water (simple diffusion model, Table 1). A reversibly-sorbed fraction is in instantaneous equilibrium with the dissolved fraction, at a ratio set by a chemical distribution coefficient. The solution of this model for a pulse-like initial input of fallout is a Gaussian distribution. However, this function is not ideal for modeling the exponential-shape profiles frequently observed in the field (Bossew and Kirchner, 2004).

Antonopoulos-Domis et al. (1995) succeeded in reproducing the long tail deep in the soil profile seen in radiocesium distributions in the field with a Diffusion-Fixation (DF) model. The model introduced an advection term for the migration of radiocesium in pore water, and a kinetic model for radiocesium binding irreversibly to a single soil binding site (Table 1). The main drawback of the model is that it does not adequately capture the gradual migration of radiocesium deeper into the ground over time.

Toso and Velasco (2001) proposed a Diffusion-Sorption-Fixation (DSF) model, which increased the number of possible sorption sites to two. It considered a reversible sorption site, with instantaneous equilibration between the dissolved and reversibly sorbed fraction, as per the simple diffusion model (Table 1). The model then introduced a second sorption site for irreversible binding of radiocesium. The kinetics of fixation were modelled explicitly using a pseudo-first order chemical reaction term. Although advective transport was neglected, the DSF model captured the exponential distribution at the terminal state. However, it could not describe the exponential distribution seen in the early stages following fallout as well as the DF model. The DSF model is similar to the two-site model proposed by van Genuchten et al. (1989) for pesticide transport and degradation in farmland soils.

The assumption of instantaneous equilibration between dissolved and reversibly sorbed radiocesium (simple diffusion and DSF models, Table 1) is limiting when trying to understand short term, dynamic, phenomena such as the depth profile immediately following fallout deposition (Bossew and Kirchner, 2004; International Atomic Energy Agency, 2009). Recently long term laboratory desorption tests of radiocesium from Fukushima soils have provided evidence of sorption sites with different kinetic rates (Murota et al., 2016). It is natural therefore to extend the DSF model with an explicit kinetic term for reversible sorption, to complement the existing kinetic fixation term, and to see the effect of different sorption rates to binding sites on the depth profile results.

This paper sets out a modified-Diffusion-Sorption-Fixation (mDSF) model where the sorption kinetics for both reversible and irreversible fixation are treated explicitly as pseudo-first order chemical reactions. A second modification is the inclusion of an advection term for the transport of radiocesium in migrating pore water. Using generic input parameters from the literature, we test the mDSF model for reproducing radiocesium depth profiles measured in Fukushima soils. The mDSF equations are solved numerically using the finite element method for spatial discretization, and the finite difference method for temporal discretization. The model is able to successfully reproduce the exponential-shape profiles with long-tails at large depths. The scope of the model is assessed by varying the input parameters within their plausible ranges. This process establishes the key parameters and their effect on the results. Finally we explore input parameters which yield radiocesium distributions displaying a peak in activity below the ground surface.

2. Methods

2.1. Model

Radiocesium can be in one of three states in the mDSF model. Mobile radiocesium is defined as the fraction that can migrate through pores in the dissolved form. Sorbed radiocesium is the fraction that is reversibly bound to soil. Fixed radiocesium is irreversibly attached to soil.

The mDSF model couples the advection dispersion equation with a fully kinetic reversible/irreversible model of sorption. Fig. 1(a) depicts the various quantities within the soil-water-air mixture, and Fig. 1(b) gives a schematic of the processes captured by the model. The radiocesium inventory obeys a mass balance equation as follows:

$$\frac{\partial A}{\partial t} + v\frac{\partial C}{\partial z} - D_e\frac{\partial^2 C}{\partial z^2} = -\lambda A, \tag{1}$$

where A (Bq m^{-3}) is the total radiocesium concentration per unit in situ volume. A is a function of the time following fallout deposition, t (s), and the depth coordinate, z (m). The parameter v (m s^{-1}) is the Darcy velocity in z-direction, C_w (Bq m^{-3}) is the mobile (dissolved) radiocesium concentration per unit volume of water, D_e (m^2 s^{-1}) is the effective dispersion coefficient for the dissolved radiocesium in the soil pores, and λ (s^{-1}) is the radioactive decay constant. The total radiocesium concentration A is