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Modeling dynamics of ^{137}Cs in forest surface environments: Application to a contaminated forest site near Fukushima and assessment of potential impacts of soil organic matter interactions

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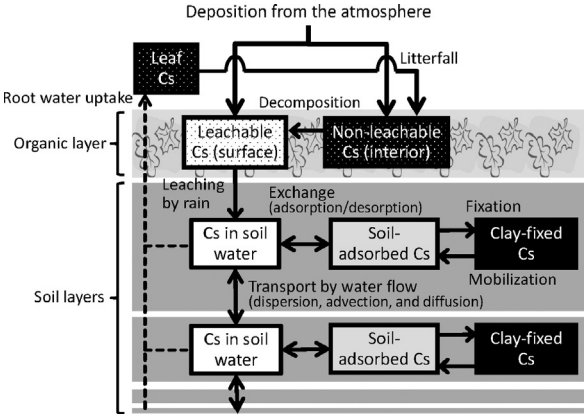
HIGHLIGHTS

- Dynamics of Fukushima-derived ^{137}Cs (Cs) in a forest was assessed by a new model
- Initially high Cs releases from the organic layer were reproduced by rain leaching
- Litter decomposition and fixation of Cs in soil controlled subsequent Cs dynamics
- Bioavailability and mobility of Cs were significantly increased in organic soils
- SOM-induced alteration of adsorption and fixation processes affected Cs dynamics

GRAPHICAL ABSTRACT



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ABSTRACT

A process-based model for ^{137}Cs transfer in forest surface environments was developed to assess the dynamic behavior of Fukushima-derived ^{137}Cs in a Japanese forest. The model simulation successfully reproduced the observed data from 3 year migration of ^{137}Cs in the organic and mineral soil layers at a contaminated forest near Fukushima. The migration of ^{137}Cs from the organic layer to the mineral soil was explained by the direct deposition pattern on the forest floor and the turnover of litter materials in the organic layer under certain ecological conditions. Long-term predictions indicated that more than 90% of the deposited ^{137}Cs would remain within the top 5 cm of the soil for up to 30 years after the accident, suggesting that the forest acts as an effective long-term reservoir of ^{137}Cs with limited transfer via the groundwater pathway. The model was also used to explore the potential impacts of soil organic matter (SOM) interactions on the mobility and bioavailability of ^{137}Cs in the soil–plant system. The simulation results for hypothetical organic soils with modified parameters of ^{137}Cs turnover revealed that the SOM-induced reduction of ^{137}Cs adsorption elevates the fraction of dissolved ^{137}Cs in the soil solution, thereby increasing the soil-to-plant transfer of ^{137}Cs without substantially altering the fractional distribution of ^{137}Cs in the soil. Slower fixation of ^{137}Cs on the flayed edge site of clay minerals and enhanced mobilization of the clay-fixed ^{137}Cs in organic-rich soils also appeared to elevate the soil-to-plant transfer of ^{137}Cs by increasing the fraction of the soil-adsorbed (exchangeable) ^{137}Cs. A substantial proportion (approximate 30%–60%) of ^{137}Cs in these organic-rich soils was transferred to layers deeper than 5 cm decades later. These results

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suggested that SOM influences the behavior of ^{137}Cs in forests over a prolonged period through alterations of adsorption and fixation in the soil.

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (hereafter, FDNPP) accident occurred in March 2011 and caused serious radioactive contamination over a wide area of eastern Japan. Among the fallout radionuclides of environmental consequence, ^{137}Cs is of primary interest because of its long half-life (30.1 years) and bioavailability associated with ecosystem nutrient cycling (Smolders and Tsukada, 2011; Hardie and McKinley, 2014). A majority (~66%) of the land area that was heavily contaminated with Fukushima-derived radiocesium (^{134}Cs + ^{137}Cs > 1 MBq m^{-2}) is covered by forests (Hashimoto et al., 2012). Contaminated forests enhance radiation exposure to the local population because of elevated ambient dose rate from the ^{137}Cs deposited on the forest surface environments (Anspaugh et al., 2002; Atarashi-Andoh et al., 2015) and through the consumption of contaminated wild vegetables (Bunzl et al., 1998; Kiyono and Akama, 2013). Therefore, predicting the behavior of Fukushima-derived ^{137}Cs after deposition is needed to understand the redistribution in the soil profile and transfer to plants in forest environments.

Processes related to forest-floor organic layers play an important role in the cycling of ^{137}Cs in forest ecosystems. In deciduous forests, the majority of the Fukushima-derived ^{137}Cs was directly deposited on the floor, because the trees were leafless in March 2011, when the FDNPP accident occurred (Koarashi et al., 2012a; Nakanishi et al., 2014). Although a significant fraction of the atmospheric ^{137}Cs was intercepted by tree canopies in evergreen forests, ^{137}Cs was shortly transferred to the forest-floor organic layers by throughfall and stemflow (Kato et al., 2012; Takahashi et al., 2015). Investigations conducted in various types of forests in the Fukushima prefecture revealed that 50%–91% of the deposited ^{137}Cs was retained in the organic layer three months after the accident (Koarashi et al., 2012a). However, a large decrease (76%) in the ^{137}Cs inventory in the organic layer between May and December 2011 was observed in a deciduous broad-leaved forest in the Ibaraki prefecture, suggesting that a faster release of ^{137}Cs occurred in the organic layer (Nakanishi et al., 2014). In contrast to these observations in Japanese forests, studies conducted in European forests have reported that a large part of Chernobyl-derived ^{137}Cs persisted in organic layers for over a decade, and ^{137}Cs retained in the organic layer is a prolonged source for the ^{137}Cs recycling in plants (Fesenko et al., 2001; Kruyts and Delvaux, 2002; Thiry et al., 2009). The observed difference in the behavior of ^{137}Cs in the organic layers may be explained by the relatively warm climate and heavy rainfall in Japan leading to faster decomposition of litter materials, and thus, thinner organic layers compared with those in many European countries (Hashimoto et al., 2013). This raises the hypothesis that since the forest-floor organic layer can act as both short-term (months to years) and long-term (years to several decades) reservoirs of deposited ^{137}Cs, storage depends upon turnover of litter materials in the organic layer under specific climatological and ecological conditions (Schell et al., 1996; Fesenko et al., 1997; Koarashi et al., 2014).

Once ^{137}Cs is transferred to mineral soil located below the organic layer, interactions with clay minerals become important in controlling the mobility and bioavailability of this radionuclide. Soil ^{137}Cs adsorbed on the specific site (i.e., frayed edge site: FES) of clay minerals (illites) has been observed to be fixed in the interlayer of the clay minerals by the collapse of the ^{137}Cs-bearing FES (Sawhney, 1972; Comans et al., 1991). This fixation results in limited redistribution (i.e., vertical migration) of ^{137}Cs by infiltrating water in the soil and reduced availability for the uptake of soil ^{137}Cs by plant roots (Smolders and Tsukada, 2011; Nakanishi et al., 2014). In the case of Fukushima, Matsunaga et al. (2013) observed that the vertical distribution of ^{137}Cs in mineral soils were almost unchanged between points in time spanning the first rainy season after the accident, which supports the very low mobility of ^{137}Cs in mineral soils in response to the rainfall events. However, Koarashi et al. (2012a) observed a deeper migration of Fukushima-derived ^{137}Cs in organic matter-rich soils (e.g., forest soils) three months after the accident, and suggested that the presence of soil organic matter (SOM) inhibits the fixation of ^{137}Cs by clay minerals by limiting the access of ^{137}Cs to the adsorption sites (Dumat et al., 2000; Rigol et al., 2002). Although SOM itself provides a high cation-exchange capacity, and is capable of attracting ^{137}Cs, the adsorption of ^{137}Cs on SOM has little selectivity and is fully reversible in comparison with other cations (Valcke and Cremers, 1994; Chibowski and Zygmunt, 2002; Rigol et al., 2002). As a result, ^{137}Cs added to organic-rich soils tends to be more mobile and bioavailable (i.e., not fixed) (Fawaris and Johanson, 1995; Kruyts and Delvaux, 2002). Forest soils in Japan generally have a high organic carbon (organic matter) content. Therefore, the influence of SOM on ^{137}Cs dynamics may be more important for Fukushima-derived ^{137}Cs in Japanese forest soils than for Chernobyl-derived ^{137}Cs in European forest soils (Koarashi et al., 2012a).

The environmental and ecological conditions observed in Japan (i.e., faster litter decomposition, heavy precipitation, and high organic matter content in soil) indicate the fate of Fukushima-derived ^{137}Cs after deposition needs to be considered for prediction of the migration processes of ^{137}Cs in Japanese forest ecosystems. A number of land-surface ^{137}Cs models have been developed since the Chernobyl accident to predict the behavior of deposited ^{137}Cs (Bergman et al., 1993; Sanzharova et al., 1994; Konoplev et al., 1993; Konoplev and Bulgakov, 1995; Schell et al., 1996; Smith and Comans, 1996; Fesenko et al., 1997; Ivanov et al., 1997; Linkov et al., 1999; Shaw and Belli, 1999; Smith et al., 1999; Almgren and Isaksson, 2006). For example, the FOA model by Bergman et al. (1993) and Sancharova et al. (1994) simulate the leaching of ^{137}Cs from the organic layer to the underlying mineral soil. The FORESTPATH model (Schell et al., 1996; Linkov et al., 1999) simulates the fixation of leached ^{137}Cs in mineral soils and the uptake of soil ^{137}Cs by standing vegetation. The RIFE1 model (Shaw and Belli, 1999) also predicts transfer of ^{137}Cs deposited on external parts of trees and forest-floor litter to mineral soil layers. As for the migration of ^{137}Cs in soil, some vertically extended models have been developed to simulate ^{137}Cs transport by infiltrating water in soil (Smith and Comans, 1996; Ivanov et al., 1997; Almgren and Isaksson, 2006). Intercomparison exercises of these models for Chernobyl forests have been extensively conducted on the framework of IAEA's BIOMASS programme (IAEA, 2002; Shaw et al., 2005). These exercises have shown that the models are subject to varying degrees of conceptual uncertainty, suggesting continued model development. After the Fukushima accident, the RIFE1 model, a relatively simple compartment model, was used to simulate the spatio-temporal dynamics of Fukushima-derived ^{137}Cs in forest ecosystems (Hashimoto et al., 2013). The modeling study predicted the time course of ^{137}Cs inventory in three forest components (tree, organic layer, and soil) in the Fukushima area. However, there have been no simulation models that completely address the underlying processes that affect the dynamic behavior of ^{137}Cs in forest soils, including ^{137}Cs transfer from the forest-floor organic layer to mineral soil; immobilization and mobilization of ^{137}Cs in the mineral soil with interactions between clay minerals and SOM; and vertical transport of ^{137}Cs by infiltrating water over the soil profile. The development of such improved models is clearly the key to accurate assessment of both short- and long-term impacts of the FDNPP accident.

In this paper, a new process-based model is presented that simulates the dynamic behavior of ^{137}Cs in forest ecosystems. The model