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Post-deposition early-phase migration and retention behavior of radiocesium in a litter–mineral soil system in a Japanese deciduous forest affected by the Fukushima nuclear accident

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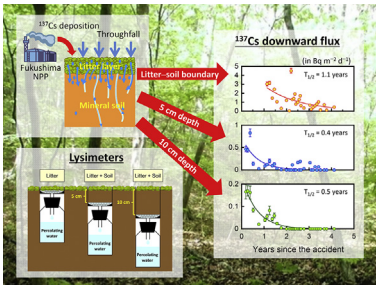
HIGHLIGHTS

- Field lysimeters were established in Japanese forest soils soon after the accident.
- Downward fluxes of ^{137}Cs were continuously monitored from May 2011 to May 2015.
- Fluxes were much greater at the litter–soil boundary and decreased with depth.
- Annual fluxes decreased year by year at all depths with seasonal variation.
- Rapid decreases in ^{137}Cs mobility reduce ^{137}Cs recycling in Japanese forests.

GRAPHICAL ABSTRACT



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ABSTRACT

The fate of radiocesium (^{137}Cs) derived from the Fukushima nuclear accident and associated radiation risks are largely dependent on its migration and retention behavior in the litter–soil system of Japanese forest ecosystems. However, this behavior has not been well quantified. We established field lysimeters in a Japanese deciduous broad-leaved forest soon after the Fukushima nuclear accident to continuously monitor the downward transfer of ^{137}Cs at three depths: the litter–mineral soil boundary and depths of 5 cm and 10 cm in the mineral soil. Observations were conducted at two sites within the forest from May 2011 to May 2015. Results revealed similar temporal and depth-wise variations in ^{137}Cs downward fluxes for both sites. The ^{137}Cs downward fluxes generally decreased year by year at all depths, indicating that ^{137}Cs was rapidly leached from the forest-floor litter layer and was then immobilized in the upper (0–5 cm) mineral soil layer through its interaction with clay minerals. The ^{137}Cs fluxes also showed seasonal variation, which was in accordance with variations in the throughfall and soil temperature at the sites. There was no detectable ^{137}Cs flux at a depth of 10 cm in the mineral soil in the third and fourth years after the accident. The decreased inventory of mobile (or bioavailable) ^{137}Cs observed during early stages after deposition indicates that the litter–soil system in the Japanese deciduous forest provides only a temporary source for ^{137}Cs recycling in plants.

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1. Introduction

The accident at the Fukushima Daiichi nuclear power plant (FDNPP) in March 2011 caused serious radioactive contamination of terrestrial ecosystems, especially forests, over a wide area of eastern Japan (MEXT, 2011a). Among radionuclides found in atmospheric fallout from the accident, radiocesium (^{137}Cs), with a half-life of 30.1 years, is the largest source of concern because of its potential impact on humans and ecosystems over the coming decades.

The fate of ^{137}Cs deposited on forest surface environments is one of the key factors in evaluating long-term radiation risks delivered from both external and internal (via plant uptake and contamination of water bodies) exposures. Immediately following the FDNPP accident, ^{137}Cs was observed to be largely retained in forest-floor litter layers (Koarashi et al., 2012a). However, in some Japanese forests, the ^{137}Cs inventory in a litter layer significantly decreased within a couple of years after deposition (Fujii et al., 2014; Nakanishi et al., 2014; Takahashi et al., 2015), suggesting that a rapid migration of ^{137}Cs occurred from the litter layer to underlying mineral soil. These observations contrast with those in European forests after the Chernobyl NPP accident, which showed that forest-floor litter layers retained the largest portion of deposited ^{137}Cs for a long time (Fesenko et al., 2001; Kruyts and Delvaux, 2002; Konopleva et al., 2009).

Once ^{137}Cs is transferred to mineral soil, interactions with soil constituents become important in controlling the mobility and bioavailability of this radionuclide. It is generally accepted that ^{137}Cs can be strongly, almost irreversibly, adsorbed by clay minerals in mineral soil layers (Sawhney, 1972), resulting in limited redistribution in the soil profile and reduced availability for uptake by plants (Sanzharova et al., 1994; Fesenko et al., 1996; Konopleva et al., 2009). There are also studies suggesting that soil organic matter has a complex effect on the mobility of ^{137}Cs in mineral soil because it offers nonspecific adsorption sites and/or because it interferes with adsorption on clay minerals (Dumat et al., 2000; Rigol et al., 2002; Koarashi et al., 2012a). Given the fact that Japanese forest soils generally have high organic matter content (Morisada et al., 2004), the influence of soil organic matter on the mobility of ^{137}Cs may be more important in controlling the mobility and bioavailability of Fukushima-derived ^{137}Cs in Japanese forest soils compared with Chernobyl-derived ^{137}Cs in European forest soils (Koarashi et al., 2012a, 2012b; Ota et al., 2016). Clearly, reasonable predictions of the fate of Fukushima-derived ^{137}Cs after deposition require an improved understanding of ^{137}Cs migration and retention behavior in Japanese forest soils under specific climatological and ecological conditions.

In the present study, we used field lysimeters to conduct four-year-long (May 2011–May 2015) observations of the vertical migration of ^{137}Cs in soils in a Japanese deciduous forest affected by the FDNPP accident. This study aimed to quantify temporal and depth-wise variations in ^{137}Cs downward fluxes over the four-year period and thus characterize the post-deposition early-phase migration and retention behavior of Fukushima-derived ^{137}Cs within the litter–mineral soil system in Japanese forest ecosystems.

2. Materials and methods

2.1. Study site

The study was conducted at two sites (termed A and B) in a temperate forest (36° 56′ N, 140° 35′ E) on an undulating plateau at the southern edge of the Abukuma mountain region (Fig. 1). The forest is located approximately 70 km southwest of the FDNPP and was affected by radioactive fallout from the FDNPP accident at a level of 10–60 kBq m^{-2} of ^{137}Cs deposition (MEXT, 2011a; Atarashi-Andoh et al., 2015). The forest is a deciduous broad-leaved forest dominated by Japanese beech (*Fagus crenata*) and Japanese oak (*Quercus serrata*). Trees at the sites had no leaves in March 2011 when the FDNPP accident occurred; therefore, it is assumed that the majority of ^{137}Cs from the fallout was directly deposited onto forest-floor litter materials (Koarashi et al., 2014; Nakanishi et al., 2014). The mean annual temperature and precipitation are 10.7 °C and 1910 mm, respectively (Mizoguchi et al., 2002). Snow cover occasionally occurs in winter up to a depth of approximately 50 cm.

Both sites are located near a mountain stream flowing through the forest; the stream has a catchment area of 0.6 km^2 (Fig. 1). Site A is the location of a previous study by Nakanishi et al. (2014), and thus, the observation results of ^{137}Cs vertical migration for the first two-year period (May 2011–March 2013) have been previously reported. Therefore, in the present study, we report results for an extended period of observation for this site, i.e., from May 2011 to May 2015. Site B is located in an area of lower elevation (approximately 600 m) within the forest catchment compared with site A (approximately 690 m). The distance between the two sites is approximately 1 km.

Late Quaternary volcanic ash has been widely deposited in this area and there is a heterogeneous and mosaic-style pattern of distribution of Cambisols and Andosols (Yoshinaga et al., 2002). At the study sites, the A horizon generally extends from the mineral soil surface to a depth of 10–15 cm, and the B horizon is located below the A horizon. The physicochemical properties of soils (0–15 cm) at each study site are presented in Table 1. In general, the properties were similar for both sites. Loam soil (classified as Cambisol) at the sites had a low bulk density (0.34–0.62 g cm^{-3}) and was acidic (pH: 4.2–5.3). The content of clay-sized particles (<0.002 mm) was less than 15% and showed no depth trend. Cation-exchange capacity (CEC) and organic carbon content were relatively higher in the top 5 cm of soil and decreased with depth.

2.2. Sample collection

Zero-tension lysimeters were installed in May 2011, two months after the FDNPP accident, to continuously collect soil water percolating through forest-floor litter and mineral soil layers at the field sites (Graphical abstract; Nakanishi et al., 2014). At each site, soil samples (litter, litter and 0–5 cm soil, and litter and 0–10 cm soil) were carefully collected using a core sampler with an internal diameter of 10 cm. The samples were then used to completely fill the lysimeters without further disturbance. Soil water percolating through the lysimeters (i.e., soil columns of different depths) was collected in 1-L borosilicate glass bottles, which were placed below the ground to maintain the samples in cool, dark conditions. Soil water in the bottles was collected roughly on a monthly basis (except in snow cover season) during the four-year period (May 2011–May 2015). Three replicate lysimeters were setup for each of the three soil samples (litter, litter and 0–5 cm soil, and litter and 0–10 cm soil) within a 2 × 2 m square (i.e., a total of nine lysimeters per site). The lysimeters for the litter sample were not installed until March 2012; therefore, ^{137}Cs vertical migration through the litter layer could not be monitored for the first year after the accident.

In the present study, we defined the periods from May 2011 to April 2012 as the first year (1Y), from April 2012 to April 2013 as the second year (2Y), from April 2013 to April 2014 as the third year (3Y), and from April 2014 to May 2015 as the fourth year (4Y). Note that 2Y differs from the period (April 2012–March 2013) that was previously defined by Nakanishi et al. (2014).