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Distribution of radiocesium in different density fractions of temperate forest soils in Fukushima

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ABSTRACT

In forested areas of Fukushima, Japan, radiocesium emitted by the Fukushima Daiichi nuclear power plant accident has accumulated on the forest floor and in surface mineral soils. To estimate and model the mobility of radiocesium in forest soils, the concept of physical variations in soil organic matter is useful. We applied density fractionation to soil samples collected in two different forest stands in Fukushima; a deciduous broadleaf forest (DBF) and an evergreen needleleaf forest (ENF). To determine the relative contributions of the two density fractions to retention of radiocesium in surface mineral soils in a temperate forest region we investigated the concentrations and stocks of radiocesium in the low- and high-density fractions (LF and HF, respectively) of the soils, as well as in the litter, and temporal changes therein between 2012 and 2014. The ^{137}Cs stock in the litter decreased from 2012 to 2014, whereas in the soil, as the sum of the LF and HF, it increased. On average, more than 40% of the ^{137}Cs stock in surface mineral soil (depth of 0–5 cm) was retained in LF particles, which were composed mainly of particulate organic matter. Although the LF accounted for only 10.3% of the dry mass of the soil on average, it was also characterized by an average concentration of ^{137}Cs that was 7.8 times higher than that of the HF, which was composed of organo-mineral particles. The mean increase in the ^{137}Cs stock in the LF from 2012 to 2014 was equivalent to 70.1% and 52.5% of those in the soils in the DBF and ENF, respectively. The relationship between loss on ignition and ^{137}Cs content indicated that the spatial heterogeneity of radiocesium in bulk soil was substantially affected by the heterogeneity in particulate organic matter. The coarse-size (> 250 μm) of the LF suggested that it was less physically protected against microbial attack. Accordingly, the LF was considered to act as a large, but temporal reservoir of radiocesium that was originally retained in the litter layer. In contrast, the accumulation of ^{137}Cs in the HF was low, especially in 2013–2014. The HF (or organo-mineral particles) might have a more important role than the LF in ^{137}Cs fixation over a long period, and should be continuously studied as part of the current radiocesium monitoring network.

1. Introduction

In forest ecosystems, the litter and soil layers act as a reservoir of anthropogenically released radioactive materials. After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in eastern Japan in March 2011, 428 km^2 of forest, representing 4% of the total forest area in Fukushima Prefecture, was contaminated with relatively high levels of radiocesium (Hashimoto et al., 2012). The concentrations and stocks of radiocesium in the components of the forest ecosystem have been monitored continuously in this region. It has been observed that in deciduous broadleaf forests (DBFs) most of the radiocesium (> 80%) was deposited on the forest floor (in the litter) and in surface mineral soil (depth of 0–5 cm) in the 6 months following the FDNPP accident (Forestry Agency of Japan, 2011). During the same period, much of the radiocesium (> 50%) in evergreen needleleaf forests (ENFs) remained in the forest canopy (Kajimoto et al., 2015; Kato et al., 2012). The rapid accumulation of radiocesium on the forest floor and in surface mineral soils necessitates estimation of the mobility of radiocesium to evaluate the future risk of radiocesium leaching into the surrounding environment and uptake by tree plantations.

Radiocesium has low mobility in surface mineral soils. Most of the radiocesium deposited following the FDNPP accident was still present in the 0–5 cm soil layer 3 years later (Forestry Agency of Japan, 2015). The clay mineral type of a soil is a factor that determines the retention

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time of radiocesium in the upper layer of mineral soils, especially in agricultural areas (Yamaguchi et al., 2012). Soil organic matter (SOM) is an important mobilizer of radiocesium in forest soils (Dörr and Münnich, 1991; Melin et al., 1994). A forest soil rich in SOM will have high radiocesium activity (Karadeniz and Yaprak, 2008; Melin et al., 1994; Takenaka et al., 1998) and decomposition of the SOM increases downward migration of radiocesium in forest soils (Dörr and Münnich, 1991; Tegen et al., 1991; Tegen and Dörr, 1996). Although previous studies have identified these close relationships between SOM content and radiocesium activity in forest soils, the positive or negative effects of SOM on the low mobility of radiocesium in surface mineral soils are unclear, partly because of variations in the form of the SOM.

The concept of physical variations in SOM (Sollins et al., 1984; Sollins et al., 1996; Wagai et al., 2013) may be useful for determining the mobility of radiocesium in forest soils. Two forms of SOM are typical: particulate organic matter (POM) and well-decomposed SOM covering mineral particles (Sollins et al., 1996; Wagai et al., 2013). POM present solely as organic matter may have a positive effect on retention of radiocesium (Rigol et al., 2002; Shand et al., 1994). The radiocesium sorbed on non-specific sites involving organic matter is retained in a more or less extractable form (Bunzl et al., 1998; Rigol et al., 2002). The POM itself is maintained by the balance of inputs and outputs of organic matter (Sollins et al., 1996) and releases radiocesium as a result of turnover. Therefore, it is possible that POM acts as a short-term reservoir of radiocesium in forest soils.

Compared with POM, the relationship between radiocesium and SOM, as an organo-mineral particle, is more complicated. Mineral particles rich in clay minerals have specific sites where radiocesium is captured, known as frayed edge sites (FESs), which contribute to fixation of radiocesium (Kruyts and Delvaux, 2002; Yamaguchi et al., 2012). Because the sorption of organic polymers by clay minerals results in decreased cesium sorption by FES (Dumat et al., 2000; Rigol et al., 2002; Valcke and Cremers, 1994), an increase in SOM-covered mineral particles may not always increase the stability of radiocesium in surface mineral soils. Accordingly, evaluation of the radiocesium content of each POM and organo-mineral particle, rather than just bulk soil, is an important step in understanding the mobility of radiocesium in forest soils.

Density fractionation has been used to separate SOM physically into different types of particles with different compositions of mineral and organic components (Shand et al., 1994; Sollins et al., 1984; Sollins et al., 1996; Toriyama et al., 2015; Wagai et al., 2013). This approach based on the specific density gradient of soil particles enables us to separate the low-density fraction (LF; i.e., SOM in the form of plant debris), which remains suspended in a heavy liquid, from the high-density fraction (HF; i.e., SOM associated with mineral particles) that settles as sediment. For example, the LF (specific density $< 1.5\ g\ cm^{-3}$) of a peaty podzol in a grassland area contaminated with radiocesium comprised 61.3% of the dry mass and was responsible for 47.8% of the radiocesium activity (Shand et al., 1994). Forest soils in Japan, as well as those in other temperate regions, are also expected to contain large amounts of LF (Parker et al., 2002; Rasmussen et al., 2005; Rovira and Vallejo, 2003), although this has not been verified. In Fukushima, where radiocesium stocks in surface mineral soils have increased (Kaneko et al., 2014), it has been suggested that the LF and HF in surface mineral soils are both responsible for retention of radiocesium. In addition, if the mineral component of the HF provides a site for the sorption of radiocesium in a stable manner compared to organic material (Shand et al., 1994), it is likely that the contribution of LF-related radiocesium to surface mineral soils decreased for a few years after the FDNPP accident, while the contribution of HF-related radiocesium increased. In this study, we considered the LF and HF to be soil particles composed mainly of POM and organo-mineral particles, respectively, although the boundary between the two is gradual because of the gradient of soil particle densities (Wagai et al., 2013).

We applied the density fractionation approach to forest soils contaminated with radiocesium in Fukushima Prefecture, Japan. To determine the relative contributions of the density fractions to retention of radiocesium in the forest soils in this region, we investigated the concentrations and stocks of radiocesium in the LF and HF of surface mineral soils, and temporal changes therein, during the 3 years after the FDNPP accident.

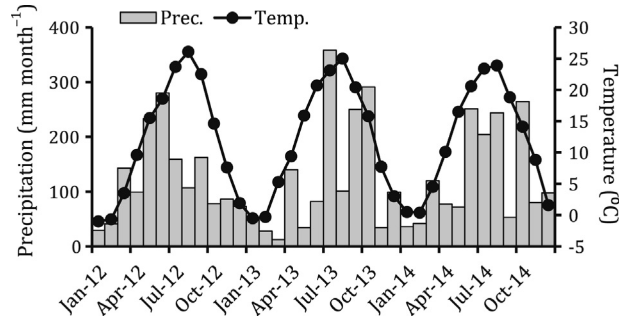

Fig. 1. Climate of the study area in 2012–2014. Monthly mean air temperatures use data for Koriyama (Japan Meteorological Agency: http://www.data.jma.go.jp/obd/stats/etrn), and monthly precipitation values use data for Tadano District (Water Information System, Ministry of Land, Infrastructure, Transport and Tourism: http://www1.river.go.jp).

2. Materials and methods

2.1. Study site and sampling

The study site was the Tadano Experimental Forest in Koriyama City, Fukushima Prefecture, which is managed by the Fukushima Prefectural Forestry Research Center. The site is a temperate forest, located about 80 km west of the FDNPP. Airborne monitoring data from 28 June 2012 indicate that the total deposition of radiocesium (^{134}Cs and ^{137}Cs) around the study site was $180\ kBq\ m^{-2}$ (MEXT, 2012). Both precipitation and air temperature are generally high in summer (Fig. 1), with a mean annual precipitation of 1503 mm (data from Tadano District for 2012–2014, Ministry of Land, Infrastructure, Transport and Tourism, http://www1.river.go.jp) and a mean annual temperature of 10.8 °C (data from the study site for 2013–2014; Hiruta et al., 2016). Sandstone and tuff form a slope with an average angle of 30° at the site and the soil moisture regime is classified as moderate in the Japanese forest soil classification system (Forest Soil Division, 1976).

DBF and ENF are the two major forest types in Fukushima Prefecture (Hashimoto et al., 2012). We selected one forest stand of each type growing at similar elevations (370–380 m) and with similar topographical features within the Tadano Experimental Forest

Table 1
Stand characteristics in Tadano experimental forest.

Variables (Unit)	Forest type	
	Deciduous broadleaf	Evergreen needleleaf
Tree density ($kN\ km^{-2}$)	81.7 (Qs) 16.6 (PP)	60.0 (Cj) 55.0 (Co)
Mean Tree height (m)	22 (Qs) 20–21 (PP)	22 (Cj) 19 (Co)
Annual litterfall (A)[a] ($kg\ m^{-2}\ year^{-1}$)	0.53	0.48
Mass of litter layer (B)[a] ($kg\ m^{-2}$)	1.88	2.13
Mean turnover of litter (A)/(B) ($year^{-1}$)	0.28	0.22

Data from Hiruta et al. (2016). Qs, *Quercus serrata*; PP, *Prunus grayana* and *Pinus densiflora*; Cj, *Cryptomeria japonica*; Co, *Chamaecyparis obtusa*.
[a] Calculated by the authors using data for 2012–2014.