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Comparison of the vertical distributions of Fukushima nuclear accident radiocesium in soil before and after the first rainy season, with physicochemical and mineralogical interpretations

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HIGHLIGHTS

► The first rainy season effect on Fukushima accident ^{137}Cs in soil was found limited.
► The ^{137}Cs relaxation lengths were almost unchanged before and after the rainy season.
► The sum of water soluble and exchangeable ^{137}Cs in soil was limited to about 10%.
► Mineralogical effects may be masked by the soil physicochemical properties.

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ABSTRACT

Effect of intense rainfall on the distribution of Fukushima-accident-derived ^{137}Cs in soil was examined. Inventories and vertical distributions of ^{137}Cs in soils were determined at 15 locations (including croplands, grasslands, and forests) in Fukushima city in the post-rainy season, approximately 4.5 months after the accident, and were compared with those in the pre-rainy season determined in our former study. The ^{137}Cs inventory levels scarcely changed between points in time spanning the first rainy season after the accident. Moreover, the majority of ^{137}Cs remained stored in the aboveground vegetation and in the upper 5 cm of soil layer at undisturbed locations in the post-rainy season. A more quantitative analysis with the characterization of the vertical profile of ^{137}Cs using the relaxation length confirmed that the vertical profile was almost unchanged at most locations. Accordingly, it is concluded that rainfall during the rainy season had a limited effect on ^{137}Cs distribution in the soil, indicating the very low mobility. Chemical extraction of ^{137}Cs from selected soil samples indicated that ^{137}Cs in the soil was barely water soluble, and even the fraction extracted with 1 M ammonium acetate was only approximately 10%. This further supports the low mobility of ^{137}Cs in our soils. Soil mineralogical analyses, which included the identification of clay minerals, suggested that smectite and mica could lower the exchangeable fraction of ^{137}Cs. However, no direct relationship was obtained between mineral composition and ^{137}Cs retention in the upper soil layer. In contrast, positive correlations were observed between ^{137}Cs extractability and soil properties such as pH, organic matter content, finer-sized particle content, and cation-exchange capacity. These results suggest that the mineralogical effect on the firm fixation of ^{137}Cs on soil constituents may be masked by the non-specific adsorption offered by the physicochemical properties of the soils.

1. Introduction

The Fukushima Daiichi Nuclear Power Station accident (IAEA, 2011; Chino et al., 2011) has posed serious concern on the fate of radionuclides in terrestrial environments. The total amount of ^{137}Cs released into the atmosphere as a result of the accident was on the order of $1–2\times10^{16}$ Bq (IAEA, 2011), and it resulted in contamination of soil with radionuclides over a wide area (MEXT, 2012; Katata et al., 2012). An aerial survey showed that as of November 1, 2011, an area of about 8400 km^2 was contaminated at greater than 60 kBq m^{-2} with ^{134}Cs and ^{137}Cs (MEXT, 2013).

The vertical distribution of radiocesium in soil is a key factor that is related to risk evaluation in terms of both external radiation exposure

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and internal exposure via plant uptake. We have studied the vertical distribution of Fukushima-accident-derived radiocesium in soil collected approximately three months after the accident (Koarashi et al., 2012a). The vertical profiles of ^{137}Cs in soils at 15 locations under different land-use types (including croplands, grasslands, and forests) were characterized by an exponential function with relaxation length. We observed a varied potential mobility of ^{137}Cs depending on the land-use types. In particular, ^{137}Cs showed a longer relaxation length in the forest soil profiles than in the cropland and grassland soil profiles, indicating a deeper penetration in the forest soil profiles. Possible factors affecting the vertical profile of ^{137}Cs included soil bulk density and soil organic carbon (SOC) content relative to clay-sized particle content.

In general, a lower bulk density provides a more highly porous soil structure. Morisada et al. (2004) reported that the mean values of the bulk density in forest soil in Japan are generally low compared with those for the world's soils in the FAO-UNESCO soil unit (Batjes, 1996). All areas of Japan, except the most northern island of Hokkaido, have a hot and very humid rainy season that lasts from early June to mid-July (the rainy season arrives about 10 days later in the northern areas including Fukushima prefecture). Most of the average annual rainfall in a region occurs in the rainy season (Japan Meteorological Agency, 2012). Such notable natural conditions observed in Japan led to the hypothesis that ^{137}Cs deposited onto and into soil might be allocated to deeper parts of the soil profile directly and indirectly by intense rainfall events during the rainy season. There is also a possibility that the rainfall events in the rainy season differently affect the migration of ^{137}Cs in soils under different land-use conditions. A lower bulk density and a higher SOC content relative to clay-sized particle content, observed mainly in forest surface soil (Koarashi et al., 2012a), might allow deposited ^{137}Cs to penetrate deeper into the soil profile owing to strong percolation of rainfall water. In contrast, a series of rainfall events might promote ^{137}Cs fixation by clay minerals via wet-dry cycles (Roig et al., 2007).

In addition to the precipitation condition, the importance of the clay mineral type for the fixation of radiocesium has been widely claimed (e.g., Facchinelli et al., 2001; Korobova and Chizhikova, 2007). For example, an increase in ^{137}Cs specific activity was observed in floodplain soil having an increased smectite content in the clay fractions (Korobova and Chizhikova, 2007). Soil organic matter may have a complex effect on the mobility of ^{137}Cs in soil because it offers non-specific sorption sites, or interferes with sorption on clay minerals (Gri et al., 2000). Ammonium acetate extraction has been frequently used to examine these effects (e.g., Askbrant et al., 1996; Rigol et al., 1999). For example, different mobility of ^{137}Cs along soil depth was clarified via the extraction method (Andersson and Roed, 1994; Bunzl et al., 1998). Thus, we believe that chemical and mineralogical analyses are imperative for understanding the dynamics of ^{137}Cs mobility under actual environmental conditions.

Since the Chernobyl accident, radiocesium fixation dynamics has become an important issue in soil system radioecology (e.g., Sanzharova et al., 1994; Absalom et al., 1995; Rigol et al., 1999). Chernobyl-accident-derived radiocesium in soil was observed to remain plant available in part despite its inherent strong association to clay minerals (Absalom et al., 1995). Sanzharova et al. (1994) reported that the mobility dynamics of ^{137}Cs, monitored by its acid soluble form, is highly dependent on soil type. These results underline the significance of field observations including soil property and mineralogical analyses. These Chernobyl-accident-related studies were conducted several years after the accident at the earliest. There is an obvious paucity of field data concerning the initial dynamics of ^{137}Cs after the fallout. Therefore, it is worth investigating possible temporal changes in radiocesium forms and associated mobility from the initial period in the present case, that is, the Fukushima accident. To our knowledge, there has been no published work on such investigations in the area affected by the Fukushima accident, with exceptions of the studies by Kozai et al. (2012) and Tanaka et al. (2012).

In this study, we compare the depth profiles of ^{137}Cs in soils at 15 locations before and after the first rainy season in 2011 to test the hypothesis—that rainfall may have an effect on the migration of Fukushima-accident-derived ^{137}Cs in soil. Soil sampling was completed during the pre-rainy season from June 18 to 20, 2011 (the first sampling), and the results of this analysis were previously reported (Koarashi et al., 2012a). In this study, a second soil sampling after the rainy season (approximately one and a half months after the first sampling) was conducted, and the depth distribution of ^{137}Cs in the soils at the same 15 locations was determined again. A sequential extraction of the soils (Andersson and Roed, 1994; Rigol et al., 2002; Vandebroek et al., 2012) and mineralogical analyses including clay mineral identification were conducted to explain changes in the depth distribution of ^{137}Cs between the two points in time spanning the rainy season (one pre-rainy season, one post-rainy season), from a physicochemical viewpoint.

2. Methods

2.1. Study site

The study site was located in the southwestern part of Fukushima city, approximately 70 km northwest of the Fukushima Daiichi Nuclear Power Station. Geological and meteorological information (parent rock, soil type, and temperature) is found in Koarashi et al. (2012a). The climate of Fukushima city belongs to a type of Dfa in the Köppen-Geiger climate classification (Peel et al., 2007). The study site was 2 km in length by 2 km in width and included one of the radioactivity surveillance points in the Deposition Density Map program (MEXT, 2012). It consists of varied land-uses and terrains, with an altitude varying between 220 and 460 m (Koarashi et al., 2012a).

In the pre-rainy season (June 18–20) in 2011, soil samples were collected at 15 locations (6 croplands, 4 grasslands, and 5 forests) within the site, and the vertical distribution of radiocesium at each location was reported by Koarashi et al. (2012a). The physicochemical properties of the soils (top 3 cm) in the investigated locations are given in Table 1 (Koarashi et al., 2012a; local soil classification also therein). In this study, we revisited these locations to collect soil samples after the rainy season. Annual rainfall has averaged 1234 mm for the last 10 years in Fukushima city, with 60% occurring between June and October. In 2011, approximately 200 mm of rainfall was recorded in Fukushima city from the day of the earthquake (March 11, 2011) to our first, pre-rainy season sampling (June 18–20, 2011). An additional 210 mm of rainfall occurred from the first sampling to our second, post-rainy season sampling.

2.2. Soil sampling

Soils were sampled at the 15 locations during the post-rainy season (July 19–20, 2011, and August 1–2, 2011). The sampling protocol was generally the same as that used in the pre-rainy season (Koarashi et al., 2012a). Soil samples were collected with a core sampler (inner diameter 9.5 cm; outer diameter 10 cm; total length 25 cm; Model HS-25, Fujiwara Scientific Company, Tokyo). One difference in the sampling protocol between the two periods is that in the pre-rainy season, aboveground vegetation (short lawn) at a grassland location (GL-1) was collected together with a soil core, and was separated in the sample treatment (see Koarashi et al., 2012a for details). The pasture grasses at three grassland locations (GL-2, GL-3, and GL-4) were mowed during the rainy season; thus, the pasture grasses collected during the post-rainy season were newly grown. Soil and vegetation samples were transported to the laboratory in a frozen state and