Chemosphere 205 (2018) 147–155

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Radiocesium distribution in aggregate-size fractions of cropland and forest soils affected by the Fukushima nuclear accident

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HIGHLIGHTS

- Influence of soil aggregation on ^{137}Cs behavior in seven soils in Fukushima.
- ^{137}Cs is largely associated with large-sized aggregates in forest soils.
- ^{137}Cs is mostly present in small-sized fractions in agricultural field soils.
- ^{137}Cs extractability is higher in large aggregates than in clay-sized fractions.
- Soil aggregation enhances the ^{137}Cs bioavailability in soils.

ARTICLE INFO

Article history:
Received 26 December 2017
Received in revised form
30 March 2018
Accepted 16 April 2018
Available online 18 April 2018

Handling Editor: Martine Leermakers

Keywords:
Radiocesium
Fukushima Daiichi nuclear power plant accident
Soil aggregates
Extractability
Soil organic matter
Clay minerals

ABSTRACT

The Fukushima Daiichi nuclear power plant accident caused serious radiocesium (^{137}Cs) contamination in soils in a range of terrestrial ecosystems. It is well documented that the interaction of ^{137}Cs with soil constituents, particularly clay minerals, in surface soil layers exerts strong control on the behavior of this radionuclide in the environment; however, there is little understanding of how soil aggregation—the binding of soil particles together into aggregates—can affect the mobility and bioavailability of ^{137}Cs in soils. To explore this, soil samples were collected at seven sites under different land-use conditions in Fukushima and were separated into four aggregate-size fractions: clay-sized (<2 μm); silt-sized (2–20 μm); sand-sized (20–212 μm); and macroaggregates (212–2000 μm). The fractions were then analyzed for ^{137}Cs content and extractability and mineral composition. In forest soils, aggregate formation was significant, and 69%–83% of ^{137}Cs was associated with macroaggregates and sand-sized aggregates. In contrast, there was less aggregation in agricultural field soils, and approximately 80% of ^{137}Cs was in the clay- and silt-sized fractions. Across all sites, the ^{137}Cs extractability was higher in the sand-sized aggregate fractions than in the clay-sized fractions. Mineralogical analysis showed that, in most soils, clay minerals (vermiculite and kaolinite) were present even in the larger-sized aggregate fractions. These results demonstrate that larger-sized aggregates are a significant reservoir of potentially mobile and bioavailable ^{137}Cs in organic-rich (forest and orchard) soils. Our study suggests that soil aggregation reduces the mobility of particle-associated ^{137}Cs through erosion and resuspension and also enhances the bioavailability of ^{137}Cs in soils.

1. Introduction

The accident at the Fukushima Daiichi nuclear power plant (NPP), triggered by a catastrophic earthquake (M9.0) and resulting tsunami on March 11, 2011, released substantial amounts of radionuclides into the atmosphere and consequently caused serious radioactive contamination of the terrestrial environment over a

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https://doi.org/10.1016/j.chemosphere.2018.04.092
0045-6535/

wide area of eastern Japan (MEXT, 2011). Of the radionuclides found in the atmospheric fallout from the accident, radiocesium (^{137}Cs), with a physical half-life of 30.1 years, is the primary source of concern because of its potential impact on humans and the environment over the coming decades.

The behavior of ^{137}Cs in soil after deposition is one of the key factors in evaluating long-term radiation risks that could be caused by both external (via elevated ambient air dose rates) and internal (via the consumption of contaminated food products) exposure. It is widely accepted that, once in soil, ^{137}Cs is strongly adsorbed (or fixed) by clay minerals (Sawhney, 1972; Cremers et al., 1988), and its mobility and bioavailability are rapidly reduced (e.g., Rigol et al., 1999; Rosén et al., 1999; Matsunaga et al., 2013; Koarashi et al., 2016b). This behavior has also been substantiated by observations that followed atmospheric nuclear weapons testing and the Chernobyl NPP accident; these observations showed that ^{137}Cs activity concentration was higher in clay-sized (<2 μm) soil particles than in the larger-sized soil particles in the surface layers of undisturbed soils (Livens and Baxter, 1988; He and Walling, 1996; Spezzano, 2005; Tsukada et al., 2008). However, there are also several lines of evidence showing that a certain proportion of ^{137}Cs in soil is not fixed and that it remains potentially mobile and bioavailable for a long time (e.g., Klioashtorin et al., 1994; Fesenko et al., 2001; Amano and Onuma, 2003; Stemmer et al., 2005). The association of soil organic matter (SOM) with clay minerals is a possible mechanism responsible for the dynamics of ^{137}Cs in soil; it may decrease the affinity of clay minerals for ^{137}Cs by blocking the specific adsorption sites, and it consequently enhances the mobility and bioavailability of ^{137}Cs because SOM itself can only offer nonspecific (easily exchangeable) adsorption sites for ^{137}Cs (Dumat and Staunton, 1999; Rigol et al., 2002; Koarashi et al., 2012a).

Soil aggregation—the binding of primary soil particles (clay, silt, and sand) together into aggregates with the help of SOM as binding agent—naturally occurs in a variety of manners and sizes in most (especially organic-rich forest) soils (Oades, 1988; Six et al., 2004). Soil aggregation depends on many factors, such as climate, vegetation, soil management practices, and soil properties, such as mineral composition, texture, SOM content, microbial activity, and moisture availability (Kay, 1998; Bronick and Lal, 2005). Of these factors, SOM particularly has a strong influence on the formation and stability of soil aggregates (Six et al., 2004).

It is well documented that soils in Japan generally have high SOM content (Morisada et al., 2004), which is attributed to the volcanic ash in soil (Nanzyo, 2002). It is therefore suggested that the influence of SOM on soil aggregation may be more important in controlling the mobility and bioavailability of Fukushima-derived ^{137}Cs in Japanese soils compared with Chernobyl-derived ^{137}Cs in European soils (Koarashi et al., 2012a; Ota et al., 2016). In addition, studies conducted in Japanese forests after the Fukushima Daiichi NPP accident have suggested a rapid migration of ^{137}Cs from the forest-floor organic layers to the underlying mineral soil (Nakanishi et al., 2014; Koarashi et al., 2016a, 2016b; Imamura et al., 2017); this highly contrasts the observations made in European forests after the Chernobyl NPP accident. Given these circumstances, reasonable predictions on the fate of Fukushima-derived ^{137}Cs after deposition rely on the better understanding of the interaction between ^{137}Cs and soil aggregates in the surface layers of mineral soil.

In this study, we investigated soil aggregation and the association of ^{137}Cs with soil aggregates in surface mineral soils affected by the Fukushima Daiichi NPP accident. Soil samples of different land types were collected at seven sites, including forests and croplands. The samples were separated into four aggregate-size fractions, and the fractions were analyzed for radiocesium to determine the ^{137}Cs distribution in the fractions. The aggregate-size fractions were also analyzed for ^{137}Cs extractability and mineral composition to explore the effects of soil aggregation on the ^{137}Cs mobility and bioavailability in the mineral soils.

2. Materials and methods

2.1. Study sites

Soil samples were collected from the upper layers of mineral soil at seven sites in Fukushima and Ibaraki Prefectures; the sites were all affected by the radioactive fallout from the Fukushima Daiichi NPP accident (Fig. 1, Table 1). Four (CP-3, CP-4, CP-6, and FR-5) of the seven sites were located in the city of Fukushima (approximately 70 km northwest of the Fukushima Daiichi NPP) and were close to each other within a 2 km × 2 km area. Since the accident in March of 2011, these sites have been extensively studied for inventory, vertical distribution, and retention of the radiocesium in soils (Koarashi et al., 2012a, 2012b; 2016a; Matsunaga et al., 2013). The CP-3, CP-4, and CP-6 sites were a leek field, a rice paddy field, and an apple orchard, respectively, and all these sites were categorized as cropland. The FR-5 site was a mixed conifer and broadleaved forest, located along a trail over a small mountain. For more details on the characteristics of these sites, including geological and meteorological information, see Koarashi et al. (2012a).

The remaining three (FR-OT, FR-NA, and FR-OG) sites were all forests dominated by deciduous broadleaved tree species, such as Japanese beech and Japanese oak. The FR-OT and FR-NA sites were in different places in Fukushima Prefecture. These two sites were selected because the soils at the sites have been classified as Andosols, which are one of the most common soil types in Japan (Takahashi et al., 2010). Andosols are highly porous, organic matter-rich soils developed from parent material of volcanic origin. The unique properties of Andosols are attributed to their well-developed aggregate structure (Nanzyo, 2002). These two sites differ in the origin and age of the volcanic deposits that produced the soil: Numazawa–Numazawako (Nm-NK) volcanic deposits with ^{14}C age of 4500–4800 years BP (before present) for the FR-OT site (Yamamoto, 2003) and Adatara (Ad-p5) volcanic deposits with ^{14}C age of 230 years BP for the FR-NA site (Yamamoto and Sakaguchi, 2000).

The FR-OG site is near the boundary between Ibaraki and Fukushima Prefectures, approximately 70 km southwest of the Fukushima Daiichi NPP. The soil at this site has been classified as Cambisols (brown forest soils), a type of soil that dominates the forest soils in Japan (Takahashi et al., 2010). This site was selected because Cambisols generally have a lower degree of aggregate formation and a less accumulation of SOM in comparison with Andosols. The FR-OG site has also been extensively investigated for inventory, spatial distribution, and vertical migration of radiocesium in soils since the accident in March of 2011 (Nakanishi et al., 2014; Atarashi-Andoh et al., 2015; Koarashi et al., 2016b). For more details on the site characteristics, see Koarashi et al. (2016b).

The inventory of ^{137}Cs in soil at the sites ranged from 10.3 to 70.9 kBq m^{-2} (Table 1). The chemical properties of the soils are given in Table 2. Soil pH was all acidic (3.5–6.1). Cation exchange capacity (CEC) ranged from 23.9 to 38.6 cmol kg^{-1}, with no remarkable difference between land-use types.

2.2. Soil sampling and treatment

Soil sampling was conducted at the four sites in the city of Fukushima in June 2011, at the FR-OT and FR-NA sites in November–December 2013, and at the FR-OG site in September 2014, respectively (Table 1). Multiple soil cores (each 10 cm in diameter and up to 20 cm in depth) were collected using a core sampler from the mineral soil surface after removing the vegetation