Science of the Total Environment 431 (2012) 392–401

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

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

Factors affecting vertical distribution of Fukushima accident-derived radiocesium in soil under different land-use conditions

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ARTICLE INFO

Article history:
Received 22 March 2012
Received in revised form 14 May 2012
Accepted 14 May 2012
Available online 15 June 2012

Keywords:
Radiocesium (^{137}Cs and ^{134}Cs)
Fukushima Dai-ichi nuclear power plant
Vertical distribution
Soil physicochemical properties
Soil organic matter
Land-use types

ABSTRACT

The Fukushima Dai-ichi nuclear power plant accident in Japan, triggered by a big earthquake and the resulting tsunami on 11 March 2011, caused a substantial release of radiocesium (^{137}Cs and ^{134}Cs) and a subsequent contamination of soils in a range of terrestrial ecosystems. Identifying factors and processes affecting radiocesium retention in these soils is essential to predict how the deposited radiocesium will migrate through the soil profile and to other biological components. We investigated vertical distributions of radiocesium and physicochemical properties in soils (to 20 cm depth) at 15 locations under different land-use types (croplands, grasslands, and forests) within a 2 km × 2 km mesh area in Fukushima city. The total ^{137}Cs inventory deposited onto and into soil was similar (58.4 ± 9.6 kBq m^{-2}) between the three different land-use types. However, aboveground litter layer at the forest sites and herbaceous vegetation at the non-forested sites contributed differently to the total ^{137}Cs inventory. At the forest sites, 50–91% of the total inventory was observed in the litter layer. The aboveground vegetation contribution was in contrast smaller (<35%) at the other sites. Another remarkable difference was found in vertical distribution of ^{137}Cs in mineral soil layers; ^{137}Cs penetrated deeper in the forest soil profiles than in the non-forested soil profiles. We quantified ^{137}Cs retention at surface soil layers, and showed that higher ^{137}Cs retention can be explained in part by larger amounts of silt- and clay-sized particles in the layers. More importantly, the ^{137}Cs retention highly and negatively correlated with soil organic carbon content divided by clay content across all land-use types. The results suggest that organic matter inhibits strong adsorption of ^{137}Cs on clay minerals in surface soil layers, and as a result affects the vertical distribution and thus the mobility of ^{137}Cs in soil, particularly in the forest ecosystems.

1. Introduction

The Fukushima Dai-ichi nuclear power plant (NPP) accident in Japan, triggered by a catastrophic earthquake (M9.0) and the resulting tsunami on 11 March 2011, caused a substantial release of radionuclides to the atmosphere (Chino et al., 2011) and subsequent radioactive contamination of the environment. Of the radionuclides found in atmospheric fallout from the Fukushima NPP accident, ^{137}Cs with a physical half-life of 30.1 years is the largest source of concern because of its impact on humans and ecosystems over the coming decades.

On 6 June 2011, the Japanese Government commenced a soil radiocesium survey to construct a soil contamination map for radiocesium around the Fukushima Dai-ichi NPP (MEXT, 2011a). In the survey, soil samples were collected from the upper 5 cm of surface soils, at one location per 2 km × 2 km grid for areas within 80 km (~2200 locations in total) from the Fukushima Dai-ichi NPP. The resulting map revealed the overall structure of the ^{137}Cs contamination field within this wide area, showing a broad range of contamination levels from less than 10 kBq m^{-2} to more than 3000 kBq m^{-2} (MEXT, 2011b). This was an important step to provide a valuable initial data on the spatial distribution of the ^{137}Cs deposition. Now strongly required are reasonable predictions of how the deposited ^{137}Cs will migrate through the soil profile and to other biological components with the passage of time, which can only be done if the processes affecting the ^{137}Cs retention or mobilization in the soils are well understood.

There are various terrestrial ecosystems in the area affected by the Fukushima NPP accident and also even in a small 2 km × 2 km area. Terrestrial ecosystems represent different ecological functions depending on their geological and ecological features (including soil characteristics), and therefore it can be expected that

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0048-9697/$ – see front matter
doi:10.1016/j.scitotenv.2012.05.041

distribution and thus mobility of ^{137}Cs in soil differ between the ecosystems or land-use types. Once in soil, ^{137}Cs is strongly bound to clay minerals (Sawhney, 1972), and its mobility in the environment driven by chemical and biological processes is very limited (Spezzano, 2005). This is evidenced by observations that ^{137}Cs was present in topsoils in a range of ecosystems even years after deposition due to the Chernobyl NPP accident (e.g., Rosén et al., 1999) and the atmospheric nuclear weapon testing (e.g. Miller et al., 1990). In contrast, other studies reported that ^{137}Cs has reached deeper the soil profile in some ecosystems. For example, Fesenko et al. (2001) found that the maximum of ^{137}Cs activity is located in different layers of forest soils, depending on the moisture regime, characteristics of litter (O horizons), and soil properties. Sanzharova et al. (1996) observed faster penetration of ^{137}Cs into the soil profile on peatlands and flooded land than on dry meadows in Russia. Because ^{137}Cs interaction in soils occurs on cation exchange sites at several levels of selectivity and reversibility, in addition to the amount and nature of clay minerals (Sawhney, 1972), other soil properties such as soil porosity, cation exchange capacity (CEC), pH, and organic matter content also can contribute to some extent to the ^{137}Cs adsorption and remobilization (Shand et al., 1994; Fesenko et al., 2001; Chibowski and Zygmunt, 2002). There are observations showing that ^{137}Cs stays available for uptake by vegetations for a longer term in organic-rich soils (Kruyts and Delvaux, 2002; Chiu et al., 2008), which has been explained by decreased specific adsorption on clay minerals by organic matter in several, direct and indirect ways (Rigol et al., 1999; Dumat et al., 2000; Rigol et al., 2002; Staunton et al., 2002). Together, these studies suggest that soil processes affecting ^{137}Cs distribution are highly complicated and strongly dependent on the ecosystems and thus the geological and ecological features there. We therefore need to investigate ^{137}Cs distribution in different soil ecosystems in our environment.

In this study, we focused on a 2 km × 2 km mesh area covering a variety of terrestrial ecosystems, located in Fukushima city affected by the Fukushima NPP accident. We collected aboveground herbaceous vegetation and forest-floor litter, and soil samples (down to up to 20 cm depth) at 15 locations within the area, and determined detailed vertical distributions of radiocesium (^{137}Cs and ^{134}Cs) inventory and of soil physicochemical properties. The first objective of this study was to characterize the vertical distribution of ^{137}Cs in soils under different land-use types (croplands, grasslands, and forests). The second objective was to identify soil properties and processes predominantly affecting ^{137}Cs retention at surface mineral soil layers across all land-use types in our site.

2. Materials and methods

2.1. Study site

The study site (2 km × 2 km mesh area) was located in the southwestern part of the Fukushima city (37.71°N, 140.36°E), ~70 km northwest of the Fukushima Dai-ichi NPP (Fig. 1). The Fukushima city is located on an extended plain to the north and south, between the Abukuma granite plateau and the Ou Mountains. Parent rocks in

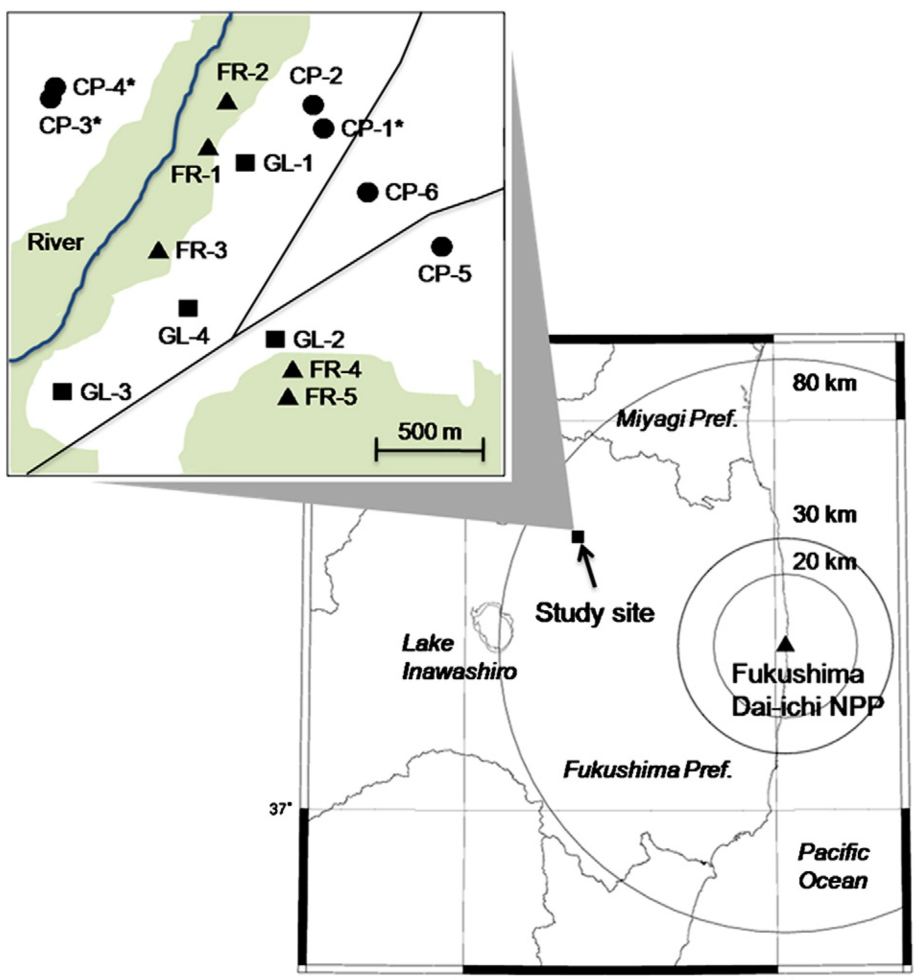

Fig. 1. Map of study area indicating 15 locations for soil sampling. Location codes CP, GL, and FR represent cropland, grassland, and forest, respectively. Notation * after location codes indicates soil disturbance.