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Short communication

Depth distribution of cesium-137 in paddy fields across the Fukushima pollution plume in 2013

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ABSTRACT

Large quantities of radiocesium were deposited across a 3000 km^2 area northwest of the Fukushima Dai-ichi nuclear power plant after the March 2011 accident. Although many studies have investigated the fate of ^{137}Cs in soil in the months following the accident, the depth distribution of this radioactive contaminant in rice paddy fields requires further examination after the typhoons that occurred in this region. Such investigations will help minimize potential human exposure in rice paddy fields. Radionuclide activity concentrations, organic content and particle size were analysed in 10 soil cores sampled from paddy fields in November 2013, 20 km north of the Fukushima power plant. Our results demonstrate limited depth migration of ^{137}Cs with the majority concentrated in the uppermost layers of soils (<5 cm). More than 30 months after the accident, between 46.8 and 98.7% of the total ^{137}Cs inventories was found within the top 5 cm of the soil surface, despite cumulative rainfall totalling 3300 mm. Furthermore, there were no significant correlations between ^{137}Cs depth distribution and the other parameters. We attributed the maximum depth penetration of ^{137}Cs to grass cutting (73.6–98.5% of ^{137}Cs in the upper 5 cm) and farming operations (tillage – 46.8–51.6% of ^{137}Cs in the upper 5 cm). As this area is exposed to erosive events, ongoing decontamination works may increase soil erodibility. We therefore recommend the rapid removal of the uppermost – contaminated – layer of the soil after removing the vegetation to avoid erosion of contaminated material during the subsequent rainfall events. Further analysis is required to thoroughly understand the impacts of erosion on the redistribution of radiocesium throughout the Fukushima Prefecture.

1. Introduction

The Tohoku earthquake and the subsequent tsunami on March 11, 2011 resulted in the Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) accident and the significant corresponding atmospheric release of radionuclides, such as ^{137}Cs ($T_{1/2}$ = 30 years) (Saunier et al., 2013). Approximately 80% of the release was transported over the Pacific Ocean with the remainder predominantly deposited on Fukushima Prefecture soils as a result of wet atmospheric fallout (Kawamura et al., 2011). Estimations of ^{137}Cs total activity in the Fukushima Prefecture soils range between 10 PBq and 50 PBq, with deposition characterized by strong spatial heterogeneities (Koo et al., 2014). The highest activities are concentrated within a 70 km long radioactive plume where initial ^{137}Cs contamination exceeded 300 kBq m^{-2} covering an area of 3000 km^2. Therefore it is crucial to understand and monitor the fate of the initial radioactive deposits in order to protect the local population against exposure to high dose rates due to gamma radiation that may prevail in areas accumulating contamination.

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In the coastal catchments affected by the FDNPP accident, Chartin et al. (2013) showed that paddy fields are one of the major sources of ^{137}Cs mobilization and export by soil erosion. A significant proportion of paddy fields are located in the upstream area of the contaminated catchments and they were shown to supply large quantities of contaminated sediment to rivers during typhoons and snowmelt events (Evrard et al., 2013, 2014). Dispersion of contamination originating from paddy fields along the rivers of the region could therefore contaminate downstream areas that were affected to a limited extent by the initial fallout. In this region, background levels of ^{137}Cs from fallout from atmospheric nuclear weapons testing were estimated to be under 100 Bq kg^{-1} (Fukuyama et al., 2005). Several studies have shown that ^{137}Cs has a low mobility in most soils (Beck, 1966; Ivanov et al., 1997) and is rapidly fixed to fine particles, especially clay minerals (Sawhney, 1972; He and Walling, 1996). These findings were confirmed in the vicinity of the main contamination plume in the Fukushima Prefecture where Saito et al. (2014) reported that ^{137}Cs was concentrated in the silt and clay fractions. Also, it was reported that the majority of ^{137}Cs remained in the first centimetre of the soil profile (Fujiwara et al., 2012; Kato et al., 2012; Koarashi et al., 2012; Lepage et al., 2014). However, it was also shown that in soils with high levels of organic matter, radiocesium may migrate down the soil profile as organic matter may reduce its affinity with clay minerals (Kamei-Ishikawa et al., 2008; Koarashi et al., 2012; Staunton et al., 2002; Szenknect et al., 2003). Koarashi et al. (2012) analysed several soil profiles sampled in the vicinity of Fukushima City and showed that clay content and Total Organic Carbon (TOC) were the main factors controlling radiocesium depth migration. They also found a strong negative correlation ($r = -0.79$, $p < 0.005$) between TOC content divided by clay content (TOC/Clay) and the percentage of retention of ^{137}Cs suggesting that the presence of organic matter inhibits the adsorption of ^{137}Cs on clay minerals.

Thirty months after the accident, it is important to investigate the depth distribution of ^{137}Cs in a selection of paddy fields located within the main contamination plume of Fukushima Prefecture. The concentration of ^{137}Cs in the uppermost layers of the soil could make it available for erosion and delivery to nearby rivers. This investigation of ^{137}Cs depth distribution in paddy fields is particularly timely in the current post-accidental phase characterised by the implementation of large-scale remediation efforts targeting paddy fields. The implications of these findings for potential soil erosion will be specifically discussed.

2. Materials and methods

2.1. Study area

The study was conducted in Fukushima Prefecture, located in North-Eastern Japan, 30 km northwest of FDNPP (Fig. 1). We focused our work on two coastal catchments (i.e. Mano and Niida River catchments − 450 km^2) draining the main part of the radioactive plume. These catchments extend from the coastal mountain range (approximately 30 km from the coast) to the Pacific Ocean, and their elevation ranges from 0 to 900 m. Mean annual rainfall was 1320 mm according to Japanese Meteorological Agency (2014) measured over 37 years at the rainfall station located in the upper part of the Nitta catchment (Fig. 1). Our study was conducted in November 2013 and cumulative rainfall reached 3300 mm (max = 35 mm h^{-1}) between the accident and our field survey (32 months) with the occurrence of 4 typhoons (Songda and Roke in 2011, Man-Yi and Wipha in 2013). In these catchments, paddy fields are predominantly located along the rivers (Tanaka et al., 2013).

Remediation works implemented since July 2012 under the supervision of the Japanese Ministry Of Environment, Government of Japan (MOE, 2012a, 2013) are concentrated in upper parts of the Nitta River catchment and consist of removing the five uppermost centimetres of the soil (Mizoguchi, 2013; Sakai et al., 2014) to decrease the radioactive dose level in order to avoid exceeding the permissible level (1 mSv y^{-1}) determined by Japanese authorities (MOE, 2012b).

2.2. Sample collection and preparation

A radiameter (LB123 D-H10, Berthold Technologies) was used to measure radiation dose rates at the ground level in the paddy fields (Table 1). To be representative, dose levels were measured at 5 different locations on each field within 10 m^2. The formula Eq. (1) proposed by the Ministry of Environment MOE (2012b) was then applied to convert these data into annual dose rates.

$$D_{an} = \frac{(D_{amb} - 0.04) * (8 + 16 * 0.4) * 365}{1000} \quad (1)$$

where D_{amb} is the ambient dose rate, and D_{an} is the annual dose rates.

Soil cores (Table 1) were collected in paddy fields across the two selected catchments (Fig. 1). We selected the fields to cover a range of dose rates (low, medium and high) in order to investigate migration of ^{137}Cs in fields with different levels of contamination (Table 1).

A soil auger (diameter 45 mm) was used to sample soil cores to a depth of 10 cm from 10 fields. The soil cores were sub-sectioned into 1 cm increments for the uppermost 5 cm (0–1; 1–2; 2–3; 3–4; 4–5 cm) and an additional 5 cm increment was taken from 5 cm to 10 cm (5–10 cm). Because of the very high radioactive dose rate measured at the location where core P9 was sampled (5.5 μsv h^{-1}), two additional layers were sampled at greater depths (10–15 cm and 15–20 cm). Density of the soil was determined by dividing the dry soil mass in each layer by its volume determined from the diameter of the soil auger and the thickness of the layer. Mean compaction in the cores was estimated to be 13% using soil density. Soil TOC content was measured with the dry combustion method (VarioTOC, Elementar, LAME, IRSN, Fontenay aux Roses, France) and clay content (particle size < 10 μm) of the samples was measured using a laser diffraction system (Malvern Mastersizer 2000) coupled to a liquid dispersing unit (Hydro 2000G, University of Paris Sud, Orsay, France). XRD measurement were performed on a diffractometer (INEL XRG 3000, L3MR, CEA, Saclay, France) equipped with a curved position sensitive detector CPS 120 and radiation were produced at 30 kV and 30 mA.

2.3. Gamma spectrometry measurements

Before measurement, samples were dried in an oven at 40 °C for a week, ground to a fine powder in an agate mortar, and then packed into 15 mL polyethylene specimen containers. Cesium-137 activities were determined by gamma spectrometry using low-background coaxial N- and P-types HPGe detectors (Canberra/Ortec). Counting times of samples varied between 80 000 s and 150 000 s. The ^{137}Cs activities were measured at the 661 keV emission peak. Counting efficiencies and energy calibration were monitored using internal, national (IRSN) and certified International Atomic Energy Agency (IAEA) reference materials (i.e., IAEA-135, IAEA-375, IAEA-CU-2006-03, IAEA-Soil-6, RGU-1, and RGTh-1) prepared in the same specimen containers as the samples. Uncertainties were estimated by combining counting statistics and calibration uncertainties. Summing and self-absorption effects were taken into account by measuring reference materials with similar densities and characteristics as the collected samples. All activities were decay corrected to the date of 14 March 2011