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^{137}Cs in irrigation water and its effect on paddy fields in Japan after the Fukushima nuclear accident

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HIGHLIGHTS

- ^{137}Cs activity in stream water was quantified under stable and storm runoff conditions.
- ^{137}Cs activity concentration increases significantly during storm runoff conditions.
- Unavailable fraction is the largest in the total ^{137}Cs activity in stream water.
- Newly added ^{137}Cs via irrigation water to inventories in soil accounts for 0.03–0.05%.
- Bioavailable fraction added via irrigation water accounts for a maximum of 3.0%.

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ABSTRACT

There is concern that radiocesium deposited in the environment after the accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011 will migrate to paddy fields through hydrological pathways and cause serious and long-lasting damage to the agricultural activities. This study was conducted in the Towa region of Nihonmatsu in the northern part of Fukushima Prefecture, Japan, (1) to quantify ^{137}Cs in stream water used to irrigate paddy fields by separating the dissolved and particulate components in water samples and then fractionating the particulate components bonded in different ways using a sequential extraction procedure, and (2) to determine the amounts of radiocesium newly added to paddy fields in irrigation water relative to the amounts of radiocesium already present in the fields from the deposition of atmospheric fallout immediately after the FDNPP accident. Three catchments were studied, and the ^{137}Cs activity concentrations in stream water samples were 79–198 mBq L^{-1} under stable runoff conditions and 702–13,400 Bq L^{-1} under storm runoff conditions. The residual fraction (F4, considered to be non-bioavailable) was dominant, accounting for 59.5–82.6% of the total ^{137}Cs activity under stable runoff conditions and 69.4–95.1% under storm runoff conditions. The ^{137}Cs newly added to paddy fields in irrigation water only contributed 0.03–0.05% of the amount already present in the soil (201–348 kBq m^{-2}). This indicates that the ^{137}Cs inflow load in irrigation water is negligible compared with that already in the soil. However, the contribution from the potentially bioavailable fractions (F1 + F2 + F3) was one order of magnitude larger, accounting for 0.20–0.59%. The increase in the dissolved and soluble radiocesium fraction (F1) was especially large (3.0% to infinity), suggesting that radiocesium migration in irrigation water is increasing the accumulation of radiocesium in rice.

1. Introduction

Substantial amounts of radionuclides have been released from the Fukushima Daiichi Nuclear Power Plant (FDNPP), which was destroyed as a result of a tsunami caused by a magnitude 9.0 Mw earthquake in March 2011. Among other radionuclides, iodine-131 (^{131}I), cesium-134 (^{134}Cs), and cesium-137 (^{137}Cs) were released after the accident and found to have been widely deposited in the environment in substantial amounts. The relatively short half-life of ^{131}I (8.05 d) means that it has already decayed to below levels that are of concern, but the longer half-lives of ^{134}Cs (2.1 y) and ^{137}Cs (30.2 y) mean that there is more potential for them to have long-lasting effects on ecosystems and agriculture in affected areas. A total of 3.58×10^{16} Bq of ^{137}Cs is estimated to have been emitted into the atmosphere from the FDNPP, and this is about 42% of the estimated amount emitted from the Chernobyl accident (Stohl et al., 2012).

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Radiocesium (later the term "radiocesium" will be used to mean ^{134}Cs + ^{137}Cs) deposition severely contaminated paddy fields in Fukushima Prefecture (Endo et al., 2013). The area of paddy fields that was contaminated with more than 5000 Bq kg^{-1} in the top 15 cm of surface soil, and was subjected to decontamination work, was estimated to be 6570 ha, and the area contaminated with more than 10,000 Bq kg^{-1} was estimated to be 2870 ha (Arita et al., 2012). About 5300 ha of paddy fields were, in 2013, still designated as restricted areas, where rice planting is strictly restricted by the central government.

The same with the primary contamination of the paddy fields from direct fallout, the secondary contamination through irrigation with contaminated water from the surrounding environment is of concern (Harada and Nonaka, 2012; Tanaka et al., 2013). Much of the radiocesium deposited in forested areas in Fukushima Prefecture is held in the canopy and in the litter layer on top of the soil (Hashimoto et al., 2012). Radiocesium is generally more mobile and easily available to plants when it is bound to organic matter (van Bergeijk et al., 1992) than when it is strongly bound to clay minerals (Almgren and Isaksson, 2006; Cornell, 1993; Shiozawa et al., 2011; Tanaka et al., 2012; Toso and Velasco, 2001). Despite its mobility, radiocesium migrates out of forest ecosystems rather slowly. Indeed, migration of less than 1% per year was found in the affected areas near the Kyshtym and Chernobyl accidents (Tikhomirov and Shcheglov, 1994). This is because radiocesium is biologically recycled within a forest ecosystem, and transport out of the ecosystem through hydrological pathways was limited in areas affected by the Chernobyl accident (IAEA, 2006).

In contrast, the transport of dissolved and particulate radiocesium through hydrological pathways is much more likely in the area affected by the FDNPP accident because there are higher levels of soil erosion, resulting from the steep topography and high levels of precipitation than in the areas in Ukraine, Belarus, and Russia that were heavily contaminated by the Chernobyl accident (Bird and Little, 2013). Some studies that were conducted in the Fukushima area after the accident showed significant increases in radiocesium concentrations in the particulate fractions in water samples taken under storm runoff conditions from close to forested areas in regions upstream of the FDNPP (Ohte et al., 2012; Yasutaka et al., 2012).

Rice planting was prohibited in 5300 ha of paddy fields in Fukushima Prefecture in 2013 to prevent contaminated rice being distributed to consumers. The Japanese government introduced a provisional limit of 500 Bq kg^{-1} for rice in 2011, but the limit was decreased to 100 Bq kg^{-1} in April 2012. Rice growing continued in paddy fields outside the prohibited areas, including those investigated in this study, but many rice farmers are concerned about the possibility of long-term migration of radiocesium from forest areas into their paddy fields in irrigation water. Indeed, paddy fields, for which large amounts of stream water are used to provide water 500–2000 mm deep during the irrigation season, are directly affected by river water quality, and are, therefore, a cause of serious concern. For example, the radiocesium concentrations in soil and rice from paddy fields irrigated with dam water were 1–1.7 times higher than that in soil and rice from paddy fields irrigated with groundwater from a well in a study in Minami-Soma, Fukushima (Endo et al., 2013).

Radiocesium was extensively monitored in river water in Fukushima Prefecture by the Forestry and Forest Products Research Institute, the Ministry of the Environment, and the MEXT. However, most of the concentrations measured by the first two organizations were categorized as "not detected". Radiocesium was successfully quantified in samples collected by the MEXT (because the method used had a detection limit of 0.1 Bq L^{-1} for both ^{134}Cs and ^{137}Cs) from 51 locations in Fukushima Prefecture between June and August 2011, and the concentrations ranged from <0.2 to 3.9 Bq L^{-1} (1.9 Bq L^{-1} ^{134}Cs + 2.0 Bq L^{-1} ^{137}Cs), with a mean of 1.12 Bq L^{-1} (0.54 Bq L^{-1} ^{134}Cs + 0.58 Bq L^{-1} ^{137}Cs) (MEXT, 2011a). Radiocesium concentrations in separated dissolved and particulate fractions of river water samples have been determined in other studies, covering relatively small areas, conducted by Ohte et al. (2012), Nagao et al. (2013), Yasutaka et al. (2012), and others. These studies have provided important information, suggesting that particulate components have played critical roles in exporting radiocesium from forest areas in the Fukushima area. However, the risks posed by river-borne radiocesium to farming have not been assessed in terms of the potential mobility and bioavailability of each component.

The study presented here was conducted (1) to quantify ^{137}Cs in stream water used to irrigate paddy fields by separating water samples into dissolved and particulate components and then fractionating the particulate components with different bond strengths using a sequential extraction procedure (SEP), and (2) to determine the amounts of ^{137}Cs newly added to paddy fields in irrigation water relative to the amounts of ^{137}Cs already present because of deposition in atmospheric fallout immediately after the FDNPP accident.

2. Materials and methods

2.1. Study fields

The study was conducted in the Towa region of Nihonmatsu, in the northern part of Fukushima Prefecture, Japan, about 40–50 km northwest of the FDNPP (Fig. 1). The mean annual precipitation and mean annual temperature between 1993 and 2012 were 1199 mm and 12.1 °C, respectively, measured at the nearest weather station operated by the Japan Meteorological Agency (Nihonmatsu Weather Station, 37°35′02″ N, 140°25′55″ E, altitude 235 m). It has been estimated, from an extensive soil survey conducted in June and July 2011, that 300–600 kBq m^{-2} of radiocesium was deposited in the study area in fallout (MEXT, 2011b). Rice has been cultivated in the Towa region continuously since the FDNPP accident, even though radiocesium concentrations of 100–500 Bq kg^{-1} were found in brown rice harvested in some of the fields in 2011 (MAFF, 2013). We studied three catchments with different land cover and land use patterns, an orchard catchment (OC), a paddy field catchment (PC), and a paddy and upland field catchment (PUC) (Fig. 2). The ^{137}Cs introduced in irrigation water was studied in a paddy field in each catchment.

2.2. Sampling

2.2.1. Water sampling

Water samples were collected from a stream adjacent to the inlet of each paddy field studied during the irrigation season, between April and September 2012. Low ^{137}Cs activity concentrations were expected in the water samples because of the results of extensive river water investigations that were previously conducted by MEXT (2011a). Samples of 500 L in stable runoff conditions and 60 L in storm runoff conditions were collected from each sampling site, each on a single occasion. The water samples were stored in plastic containers in cool dark conditions and wrapped to avoid exposure to direct sunlight, for a maximum of one month. No chemical or medicinal substances were added to prevent the growth of microorganisms. The water samples during storm runoff conditions were collected on 20 June 2012, around when the precipitation rates were 66 mm d^{-1} in the OC and 55 mm d^{-1} in the PC and the PUC.

2.2.2. Paddy field soil sampling

Soil samples were collected from each of the paddy fields that were studied on 1 May 2012, immediately before the irrigation season started. Three sampling points were selected at each study site, and they were (1) within 3 m of the water inlet, (2) in the center of the field, and (3) within 3 m of the water outlet. A soil core was collected from the plowed layer (ca. 0–15 cm deep) at each sampling point using a soil sampler (5 cm i.d. × 30 cm long; HS-30; Fujiwara Scientific Company, Tokyo, Japan) and thoroughly mixed.