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Temporal changes of radiocesium in irrigated paddy fields and its accumulation in rice plants in Fukushima

Baolu Yang [a, b, *], Yuichi Onda [b], Yoshifumi Wakiyama [c], Kazuya Yoshimura [d], Hitoshi Sekimoto [e], Yiming Ha [a]

[a] *Institute of Agro-products Processing Science and Technology, Chinese Academy of Agricultural Sciences, Beijing, 100193, China*
[b] *Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, Tsukuba, 305-8572, Japan*
[c] *Institute of Environmental Radioactivity, Fukushima University, Kanayagawa 1, Fukushima, 960-1296, Japan*
[d] *Sector of Fukushima Research and Development, Japan Atomic Energy Agency, Tokyo, 100-8577, Japan*
[e] *Faculty of Agriculture, Utsunomiya University, Utsunomiya, 321-8505, Japan*

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ABSTRACT

About half of the total paddy field area, which is the dominant agricultural land in Fukushima Prefecture, was contaminated by radiocesium released by the Fukushima Daiichi Nuclear Power Plant accident. In this study, we investigated the temporal changes of radiocesium in soil, irrigation water, and rice plant in two adjacent rice paddies, with and without surface-soil-removal, in Fukushima Prefecture for over three years (2012–2014) after the nuclear accident. Our results showed that radiocesium migrated into 24–28 cm soil layers and that the activity concentration of radiocesium in paddy soils showed a significant reduction in 2014. The newly added radiocesium to paddies through irrigation water contributed only a maximum value of 0.15% and 0.75% of the total amount present in control and decontaminated paddies, respectively, throughout the study period. The radiocesium activity concentration in suspended sediment in irrigation water exponentially decreased, and the effective half-lives (T_{eff}) for ^{137}Cs and ^{134}Cs were 1.3 and 0.9 years, respectively. Additionally, the average suspended sediment concentration in irrigation water increased between 2012 and 2014, suggesting that enhanced soil erosion had occurred in the surrounding environment. Radiocesium accumulation in rice plant also decreased with time in both paddies. However, the concentration ratio of radiocesium for rice plant in the decontaminated paddy increased compared with control paddy, despite approximately 96% of fallout radiocesium removed in paddy soil. Further analysis is required to clarify the reasons of high concentration ratio of radiocesium for rice plant in the decontaminated paddy.

1. Introduction

On March 11, 2011, the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred as a consequence of a big earthquake followed by a tsunami. The accident released a substantial amount of radionuclides into the environment. Among the radionuclides emitted from the accident, radiocesium (^{137}Cs and ^{134}Cs) is a primary concern because of its long half-lives. International Atomic Energy Agency (IAEA, 2012) estimated that the ^{137}Cs released into the environment from the disastrous nuclear accident was approximately 8.2 PBq. The region most affected by the nuclear accident is Fukushima prefecture, which is dominated by rice paddies. The accident left about 453 km^2 of paddy fields (i.e., 43.1% of the total paddy fields) contaminated by radiocesium. The soil contamination by radiocesium exceeding 5000 Bq/kg accounted for 5.9% of the investigated paddy fields with an area of 61 km^2 (Wakahara et al., 2014). According to a survey, the highest content of radiocesium in the paddy soil was 41,400 Bq/kg (Bachev, 2015).

Cesium is an alkaline element, and given its similarity with the nutrient element potassium, radioactive cesium is readily absorbed into edible and nonedible parts of rice grown in contaminated farmlands (Smolders and Tsukada, 2011; Kobayashi et al., 2014). Following the Fukushima nuclear accident, the activity concentration of radiocesium in brown rice which was produced in some paddy fields in Fukushima prefecture exceeded the Japanese

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* Corresponding author. Institute of Agro-products Processing Science and Technology, Chinese Academy of Agricultural Sciences, Beijing, 100193, China.
E-mail address: yangbl2011@hotmail.com (B. Yang).

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provisional safety limit value of 500 Bq/kg (Saito et al., 2012). For instance, 1050 Bq/kg radiocesium was reported in cesium-contaminated rice samples taken in the region 50 km away from the Fukushima Daiichi reactors (Bachev, 2015). Moreover, beef was also contaminated by cesium, because the animals were fed contaminated rice straw (Bachev, 2015). Ingesting food contaminated with radiocesium could then pose health risks.

To assure food safety, 11,200 ha of paddy fields where the radiocesium activity concentration exceeded the limit of 5000 Bq/kg were mandatorily restricted for planting by the Japanese government in April 2011 (Bachev, 2015). The guideline level for radiocesium in crops was also decreased to provisional 500 Bq/kg in March 2011, and then to 100 Bq/kg in April 2012 (Miyashita, 2014; Yoshikawa et al., 2014). Simultaneously, the Japanese government has performed various decontamination methods, such as surface-soil removal, inverting plowing, and padding with water, according to the activity concentration of radiocesium in the farmland soil (MAFF, 2011). Surface-soil removal reduced 80%–90% of the radiocesium level in the farmland soil and 60%–80% of the air dose rates (MAFF, 2013). Sakai et al. (2014) reported that soil removal also reduces the activity concentration of radiocesium in tadpoles. However, despite surface-soil removal being the major method used to decontaminate the Chernobyl accident site (Vovk et al., 1993), there is little documented quantitative information about the effect of surface-soil removal on radiocesium accumulation in rice plants because it is the first time that the rice paddy was seriously contaminated by radioactive materials.

Rice is usually flooded with 5–15 cm of water during the crop cultivation season (IAEA, 2009). As well as the contamination of the paddy fields from direct radiocesium fallout, contamination through contaminated irrigation water needs to be considered. Recent studies have reported that 84–92% of the total radiocesium in the fluvial system was transported via particle form and absorbed in the suspended sediment of river water (Yamashiki et al., 2014). If irrigation water is coming from a contaminated river, then suspended sediments (SS) containing radiocesium will accumulate in paddy fields. Some studies reported that the added radiocesium through irrigation water is negligibly small compared with the originally contained radiocesium in non-decontaminated paddy field (Tanaka et al., 2013; Yoshikawa et al., 2014). However, Sakai et al. (2014) reported that the activity concentrations of radiocesium in topsoil became 3.8 times higher in a decontaminated paddy one year after surface-soil removal because of irrigation water and atmospheric deposition. Four years after the FDNPP accident, no further reports have been identified by the authors with regard to the environmental behavior of radiocesium in decontaminated paddies.

In this study, we investigated the vertical distributions and temporal changes of radiocesium in the soil profiles of decontaminated and non-decontaminated paddy fields from 2012 to 2014. We also discuss the accumulation of radiocesium through irrigation water and simulate variation in radiocesium activity concentration in SS from irrigation water. In particular, we focus on the effect of surface-soil removal on radiocesium accumulation in rice plants.

2. Materials and methods

2.1. Study site

This study was conducted in Yamakiya District of Kawamata Town, Fukushima Prefecture, which is about 40 km northwest of the FDNPP (Fig. 1). The mean rainfall was 836 mm during the rice cultivation season (May to October) from 1999 to 2010, measured at the Yamakiya weather station operated by the Japan Meteorological Agency.

Two adjacent lowland rice paddies were selected as test fields (Fig. 2). One paddy was decontaminated (520 m^2), in which the surface soil (5–10 cm) was removed on June 12, 2011, after the Fukushima nuclear accident; the other paddy remained as an unmodified control paddy (510 m^2). Rice paddies are normally enclosed with low embankments that can maintain 5–10 cm of still water in the fields. The irrigation water of both paddies was supplied from the same drainage basin that originated from the Kuchibuto River. As the irrigation water input for the two paddies depended on many different factors including the seasonal heavy rains, and some other uncontrollable factors like the damage of paddy embankments, etc., to best meet the requirements of water in different growth period of rice plants, the water input amount at a time and the irrigation frequency were determined based on the actual situation of water in each paddy during the cultivation season. Therefore, the total amount of irrigation water input was not artificially controlled to remain the same for each paddy in different years, and between the two paddies in the same year.

Two rice cultivars (*Oryza sativa* L. cv. Koshihikari and Hitomebore) were planted for four consecutive years, from 2011 to 2014, respectively in the two plots, as shown in Fig. 2, according to the local farming time and procedures. This study only discusses the data from 2012 to 2014 because some data in 2011 were not available. To evaluate the load of SS and radiocesium in the irrigation water during the cultivation season, the rainfall, irrigation water flow rate, and activity concentrations of SS and particle radiocesium were monitored. The water flow rate was determined using a three-inch Parshall flumes combined with a water level gauge (WT-HR, TruTrack, ChristChurch, New Zealand) based on the relationship of water level height with manually measured discharge. To determine whether the changes of water level were caused by irrigation water or rainfall, the rainfall was measured using a rain gauge (Rain Collector II, Davis Instruments, Hayward, CA, USA). The activity concentration of SS in the irrigation water was monitored using a turbidimeter (ANALITE NEP180/30G, McVan Instruments, Victoria, Australia). The data obtained by the water level gauge, rain gauge, and turbidimeter were recorded automatically every 10 min by a data logger. The load of SS from irrigation water was calculated based on the water discharge and the sediment density in the water. In addition, a time-integrated SS sampler (Phillips et al., 2000) was installed to collect the SS transported by water which was used to measure the radiocesium activity concentration. The specific test field conditions are shown in Fig. 2.

2.2. Sample collection and preparation

2.2.1. Paddy soil and suspended sediment of irrigation water

Removal of 5–10 cm surface-soil in the experimented paddy was conducted on June 12, 2011. To obtain the initial inventory of radiocesium, five surface soil samples with a depth of 0–5 cm were collected from each paddy field on June 5, 2011 using the method of Onda et al. (2015), after the removal of surface soil, the soil sampling was repeated on June 12, 2011. The soil type was classified as gley soil with a fine texture in the upper surface horizon, and the grain size distribution and organic matter content were similar between the two paddies (NIAES, 1995; Sakai et al., 2014). After sampling, both paddies were cultivated for rice planting.

Soil core samples of 30 cm depth (φ50 mm) were respectively collected at 12 sites on April 20, 2012, October 26, 2012, May 25, 2013 and November 1, 2014 in the two paddies (Fig. 2), while only 10 soil core samples with a depth of 30 cm from each paddy were collected on November 1, 2014. The soil core samples were separated into seven positions at an interval of 4 cm from the surface. Samples were oven-dried at 105 °C for 16–24 h, and then ground to <2 mm. After homogenization, an aliquot of each soil core sample