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Soil radiocesium distribution in rice fields disturbed by farming process after the Fukushima Dai-ichi Nuclear Power Plant accident

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A B S T R A C T

A magnitude 9.0 earthquake and subsequent large tsunami hit the northeastern coast of Japan on March 11, 2011. This resulted in serious damage to the reactors of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), operated by the Tokyo Electric Power Company. Large amounts of radionuclides were released from the FDNPP, a proportion of which were deposited onto the ground. In this study, we investigated soil radiocesium contamination of rice fields in Aga and Minamiuonuma, Niigata, ~130 and 200 km away from the FDNPP, respectively, as Niigata is one of the largest rice growing regions in Japan. Soil samples were collected from the plow layer of five rice fields in August and September, 5–6 months after the FDNPP accident. Results showed that radiocesium concentrations (the sum of Cs-134 and Cs-137) in the rice soil samples were ~300 Bq (kg dry soil)$^{-1}$. All samples contained a Cs-134/Cs-137 activity ratio of 0.68–0.96 after correction to March 11, 2011, showing that the radiocesium released from the FDNPP were deposited on these areas. Although the rice fields had been disturbed by farming processes after the FDNPP accident, the depth distribution of radiocesium concentrations in the plow layers showed higher concentrations in the upper soil layers. This suggests that spring tillage, flooding and puddling performed before rice transplantation may not disperse radiocesium deposited on the surface through the whole plow layer. In addition, the planar distribution of radiocesium concentrations was examined near the water inlet in one of the rice fields. Highest activities were found aligned with the direction of irrigation water discharge, indicating that radioactivity levels in rice fields may be elevated by an influx of additional radionuclides, probably in irrigation water, during farming.

1. Introduction

On March 11, 2011, a magnitude 9.0 earthquake occurred 130 km off the Pacific coast of the Tohoku region of Japan (Japan Meteorological Agency, 2011). The quake struck a wide area of northeastern Japan and was followed by a large tsunami. This resulted in serious damage to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), operated by the Tokyo Electric Power Company (TEPCO). The entire FDNPP plant was flooded and the cooling system of the nuclear reactors failed. Three nuclear reactors experienced full nuclear meltdown and pressures inside the reactors mounted. Although TEPCO attempted to vent this pressure to the atmosphere, hydrogen explosions occurred (Dauer et al., 2011). Japanese authorities have declared the accident to be level 7, the highest level on the International Nuclear Event Scale rated by the International Atomic Energy Agency (Ministry of Economy, Trade and Industry, Japan, 2011). As a result of the accident, large amounts of radionuclides were released into the atmosphere and the ocean (Chino et al., 2011; Nuclear Safety Commission, 2011). Radio xenon, I-131, I-132, Te-132, Cs-134, and Cs-137 have been detected not only in Japan, but also in other countries, and have circulated around the globe (Lozano et al., 2011; Manolopoulou et al., 2011; Momoshima et al., 2011).

The Nuclear Emergency Response Headquarters, Government of Japan (2011) estimated that the total amounts of I-131 and Cs-137 released into the atmosphere from the FDNPP were approximately 1.6×10^{17} and 1.5×10^{16} Bq, respectively. More recently, Stohl et al. (2011) reported that inversion results gave a total emission of 3.58×10^{16} Bq for Cs-137. Of the total Cs-137 emission from the crippled nuclear power plant, about 20% is thought to have been deposited onto Japanese land (Morino et al., 2011; Stohl et al., 2011), resulting in serious soil contamination in Fukushima and the surrounding prefectures (Ministry of Education, Culture, Sports, Science and Technology, Japan, 2011). Yasunari et al. (2011) estimated that the soils around the FDNPP have been contaminated with depositions of more than 100×10^9 Bq km^{-2} and those of neighboring prefectures with 10×10^9 Bq km^{-2}.

The fallout also caused widespread radionuclide contamination of farmlands, mainly in the eastern part of Japan. The Ministry of Agriculture, Forestry and Fisheries created a map of radiation levels in farmlands based on analysis of soil samples collected from about 580 locations in the prefectures of Fukushima, Miyagi, Ibaraki, Tochigi, Gunma and Chiba (Ministry of Agriculture, Forestry and Fisheries, Japan, 2011). It shows that farmlands severely contaminated with >5,000 Bq (kg dry soil)$^{-1}$ of radiocesium were found ~50 km northwest of the FDNPP.

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Moderately contaminated agricultural lands containing 1,000–5,000 Bq (kg dry soil)$^{-1}$ were mainly distributed around the area above, and in the Naka-dori region of Fukushima Prefecture and a northern part of Tochigi Prefecture. Radiocesium levels in the other soils were <1,000 Bq (kg dry soil)$^{-1}$, including from several locations bordering Niigata Prefecture.

Japanese farmland affected by radionuclide deposition from the FDNPP includes large areas of rice paddies. Since rice is the most important staple food in Japan, radionuclide contamination in rice paddies and its transfer into rice grain are a major concern. Niigata, adjacent to Fukushima Prefecture, is one of the largest rice-growing prefectures, where rice production and rice-related industries are major industries. Although it is thought that 0.2% of the total Cs-137 emission was deposited in the Niigata Prefecture (Morino et al., 2011), few figures on soil radionuclides in rice fields collected in the Niigata area have been available (Niigata Prefectural Department of Agriculture, Forestry and Fisheries, 2011a, 2011b).

The present study investigated contamination levels of soil radionuclides in rice fields in Niigata, 5 to 6 months after the FDNPP accident. The effects of usual rice farming management were also examined by measuring the depth and planar distribution of radionuclides in the plow layer. According to Tagami et al. (2011), Te-129 m, I-131, Cs-134, Cs-136 and Cs-137 were largely detected in soil samples collected from ~20 km south of the FDNPP after the accident. The effects of Cs-134 and Cs-137 on environments can be more problematic in the long term, since the two radionuclides have half-lives of 2 and 30 years, respectively, which are longer than other radionuclide half-lives. These radionuclides were therefore the focus of this study.

2. Materials and methods

2.1. Study site

After the FDNPP accident, the Niigata prefectural government began monitoring radiation dose rates in air at 1–13 m above ground level with portable radiation monitoring equipments (Ohyo Koken Kogyo, Fussa, Japan) at six points from March 12 (Niigata Prefectural Government, 2011). Results showed that air radiation dose rates rose sharply on March 15, especially at monitoring posts installed in Minamiuonuma City in the Chuetsu region and Aga Town in the Kaetsu region (Fig. 1). Maximum air radiation dose rates at Minamiuonuma and Aga were 0.527 and 0.230 μSv h^{-1}, respectively. Air radiation dose rates then gradually decreased. The monitoring results suggest that weather conditions transferred non-negligible amounts of airborne radionuclides from the FDNPP to Niigata Prefecture, which then fell to the ground in rain or snow (Ioannidou and Papastefanou, 2006), resulting in soil contamination in the parts of Niigata Prefecture. Therefore, soil samples were taken from rice fields in Aga and Minamiuonuma in this study (Fig. 2). These are located in areas that border Fukushima and Gunma prefectures. Daily and cumulative precipitation near the sampling sites during the period of 11–31 March, 2011 is shown in Fig. 3.

The two rice fields in Aga (A1 and A2) were ~130 km west-northwest of the FDNPP. They were ~0.5 km apart and separated by a tributary of the Agano River. The four rice fields in Minamiuonuma (M1–M4) were ~200 km west-southwest of the FDNPP. Sites M1–M3 were in the Uono River basin, and site M4 was in the Nobori River basin. Sites M2, M3 and M4 are 3, 0.3 and 6 km away from site M1, respectively.

Rice cultivation had followed normal soil management practice in the all rice fields. While cultivation schedules in 2011 varied with each field, they can roughly be summarized as follows: spring tillage, flooding and puddling in May, transplantation in May–early June, midsummer drainage at the end of June–July, and harvest in September–October.

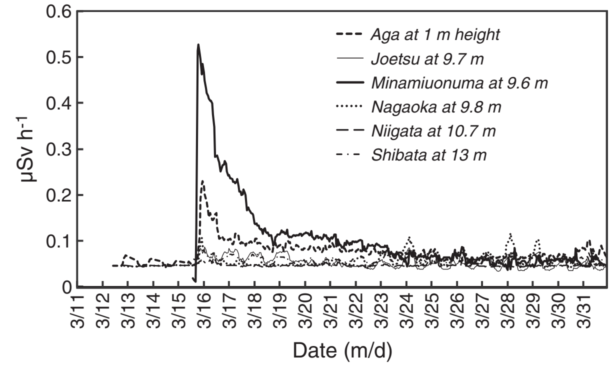

Fig. 1. Radiation monitoring results from March 12 to March 31, 2011, at six monitoring posts installed in Niigata Prefecture after the FDNPP nuclear accident (each location is presented in Fig. 2). External radiation dose rates were measured with portable radiation monitoring equipments at 1–13 m above ground level, expressed in terms of dose rates in air. Illustrated by the authors based on Niigata Prefectural Government (2011).

2.2. Sampling

Soil samples were collected from the plow layer (0–20 cm) at sites A1, A2, M1 and M2 on August 23, 2011, to determine the levels of radiocesium contamination. The rice fields had been drained, and no covering water was observed. Three sampling points, point A within 3 m of the water inlet, point B in the center of the rice field, and point C within 3 m of the drain outlet, were selected for each site. Soil samples were collected from the plow layer (0–20 cm depth) at each sampling point using a hand trowel and thoroughly mixed. Rice straw, ears and grain were also collected from the sites.

To investigate the depth distribution of radionuclides, soil samples were collected from site M1 on September 6 and sites M3 and M4 on September 20 at points A, B and C as above. All rice fields had been drained. Four 5-cm diameter soil cores of 0–13 cm depth for site M1 and 0–15 cm depth for site M3 and M4 were randomly collected at each sampling point. Each core was sectioned into three parts (0–5, 5–10 and 10–13 cm for site M1, and 0–5, 5–10 and 10–15 cm for M3 and M4), and each depth section was thoroughly mixed.

The planar distribution of radionuclides was investigated around the water inlet at site M1 on September 20. Twelve grid-like sampling points were selected as shown in Fig. 4. Four 5-cm diameter and 5-cm depth soil cores were collected from the surface layer of each sampling point, and mixed well.

2.3. Radiocesium measurement

Radiocesium in the soil and plant samples was analyzed by the Niigata Environment Hygiene Central Laboratory Co. (Nagaoka, Japan). Part of each soil sample was dried to constant weight at 105 °C (12–36 h), and coarse organic matter and stones were removed by hand. Following this, the soil sample was crushed using a Forcemill FM-1 (Osaka Chemical Co., Osaka, Japan) and ca. 100 g was transferred to a 127 mL U-8 polystyrene cylindrical bottle (external size: 5 cm diameter × 6.8 cm height). Concentrations of Cs-134 and Cs-137 were determined with a GC3018 Ge gamma-ray detector connected to a multichannel analyzer system (Canberra Industries Inc., CT, USA). The gamma spectra obtained were analyzed with Gamma Explorer (Canberra Industries Inc.). A true coincidence summing correction considering the container geometry was applied. Gamma-ray emissions at energies of 604.66 and 661.64 keV for Cs-134 and Cs-137, respectively, were counted for 1,800–7,200 s to secure each 10 Bq (kg dry soil)$^{-1}$ as quantitative limits for Cs-134 and Cs-137 calculated using the method of Cooper (1970). Nine nuclide mixed activity standard volume sources in alumina (Japan Radioisotope Association, Tokyo, Japan) were used as reference standards.