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ARTICLE

Behavior of radiocesium in decontaminated paddy fields in Fukushima Prefecture, Japan

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Abstract

The 2011 Great East Japan earthquake and tsunami led to dispersal of radionuclides into the surrounding area from the Tokyo Electric Power Company's Fukushima Daiichi Nuclear Power Plant. Before agricultural activities can safely resume after this accident, it is necessary to evaluate the dynamics of radiocesium in irrigation water in the area. We measured the concentrations of total and dissolved radiocesium in irrigation water and analyzed radiocesium concentrations in rice and soil samples, the soil to brown rice transfer factor (TF) in two decontaminated paddy fields located within the zone where paddy rice culture was prohibited, and radiocesium/water balance in a paddy field in 2014. Our key findings were as follows: (1) about 85% of the radiocesium in the irrigation water did not drain away but accumulated in the paddy field; (2) in comparison with the amount of the radiocesium deposited in the fields, the amount of radiocesium that accumulated during irrigation was approximately 0.076%; (3) the outflow of total radiocesium from paddy field was 13.0% of the inflow to a paddy field (irrigation water and fallout); (4) the soil to brown rice TF was 0.0015–0.0068 in the decontaminated paddy fields where soil improvement was performed to increase the content of exchangeable potassium to 200 (mg K)/kg soil before the conventional application; and (5) the concentration of radiocesium in brown rice was about 2% of the standard limit in Japan (100 Bq/kg), and the impact on brown rice from radiocesium in the irrigation water was limited.

Keywords Paddy field · Irrigation water · Radiocesium · Fukushima · Decontamination · Radionuclide

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Introduction

On March 11, 2011, the Great East Japan earthquake and tsunami caused an accident at the Tokyo Electric Power Company's Fukushima Daiichi Nuclear Power Plant (FDNPP). The radionuclides released and dispersed by this accident were deposited mainly in Fukushima Prefecture (Endo et al. 2012; MEXT 2011). The total area of farmland contaminated with radiocesium concentrations > 5000 Bq/kg in the plow layer (the upper 15 cm of soil) was estimated at approximately 8900 ha (MAFF 2012a). Paddy field cropping restrictions were imposed in the area in 2011 to limit the transfer of emitted radionuclides to agricultural crops (MAFF 2014b).

Various decontamination measures, such as topsoil removal, the use of inverting plows, and decontamination using soil puddling with water, were implemented in the paddy fields and irrigation and drainage canals (MAFF 2012b). The decontamination work of the government's planned agricultural land (31,060 ha) was completed at 99.9% as of February 2018 (MOE 2018).

Tsukada et al. (2002) found that the geometric mean of transfer factor of ^{137}Cs from paddy soil to polished rice was 0.0016, its 95% confidence interval was 0.00021–0.012, and the transfer factors of ^{137}Cs decreased with increasing potassium concentration in the paddy soil. Analysis of the factors affecting the transfer of radiocesium from the paddy field soil to rice crop in 2011 revealed that the content of exchangeable potassium in the soil played an important role (Kato et al. 2015). Potassium fertilization effectively reduced radiocesium uptake due to the antagonism between potassium and radiocesium, with little or no effect on rice production (Eguchi et al. 2015; Fujimura et al. 2016a). Therefore, soil improvement to increase the content of exchangeable potassium to 200 (mg K)/kg soil before the conventional application was proposed as a countermeasure against radiocesium absorption (Kato et al. 2015).

Recommended measures to control of radiocesium absorption (for example, by potassium fertilization) contributed to reduction of the concentration of radiocesium in brown rice (Ota 2014). The results of the monitoring studies revealed that, at several sites, the radiocesium concentration in environmental water (river, lake, and groundwater) since the FDNPP accident was less than the quantification limits or 1 Bq/L (MAFF 2013a; MOE 2012a, b). To date, in areas where cropping has recommenced, there have been no confirmed reports of the effect of radiocesium in irrigation water on crops.

However, before farming can recommence in areas such as the restricted residence zone, the impact of radiocesium on farming must be investigated. In particular, there are concerns about the transfer of radiocesium to irrigation water sources such as rivers and ponds via surface runoff and the consequent emergence of new hot spots of contamination in agricultural irrigation facilities (Kubota et al. 2014; Yoshikawa et al. 2014).

Radiocesium in environmental water exists not only as dissolved ions, but also as suspended particulates, which are adsorbed or fixed by suspended organic and inorganic particles (MAFF 2014a). Tsukada et al. (2017) reported the geometric mean of Kd, determined from 54 irrigation waters in 2014 in the Fukushima Prefecture, was 110,000 over one order of magnitude higher than that reported by IAEA (2010) from 29,000 in freshwater ecosystems determined by field experiments. A higher Kd is associated with the higher the distribution of radiocesium primarily in the solid phase in the equilibrium state; i.e., at the same radiocesium concentration, the concentration of dissolved radiocesium is lower. Suspended radiocesium is not easily dissolved; therefore, its absorption by rice in paddy fields is low. In contrast, dissolved radiocesium can be directly absorbed by plants and is thought to transfer easily to brown rice (MAFF 2014c; Suzuki et al. 2015). On the other hand, Shiozawa (2012) suggested that the contamination of brown rice in 2011 mainly occurred because of fallout deposited on the organic matter covering the paddy fields and not via irrigation water. It is important to elucidate the impact of radiocesium in irrigation water on brown rice before agricultural activities recommence. This task involves clarifying the changes over time in the radiocesium concentration, quantifying the concentration of radiocesium, and determining the behavior of radiocesium in irrigation water in paddy fields.

In the present study, we conducted trials in decontaminated paddy fields at a site in a zone with restricted cropping. Our objectives were to (1) quantify the mass balance of radiocesium in a paddy field by analyzing the water balance; (2) investigate the behavior of radiocesium in irrigation water in decontaminated paddy fields; and (3) clarify the effect of the radiocesium in irrigation water on the radiocesium concentration in brown rice.

Materials and methods

Study area and sampling sites

We conducted our research in two decontaminated paddy fields at site *K* which was in the "restricted residence zone," designated in March 2014, and located ca. 40-km northwest of FDNPP in Fukushima Prefecture, Japan (Fig. 1).

In 2012, the Ministry of Agriculture, Forestry and Fisheries (MAFF) performed topsoil removal at the site *K* as a test case, and decontamination at this site *K* was confirmed. From 2012 to 2014, paddy field rice and vegetables were grown in cultivation trials to measure the transfer of radiocesium to crops through irrigation water after decontamination (MAFF 2012c, 2013b). In the present study, we assessed the quality of the irrigation water and analyzed the results of cultivation trials conducted in 2014. To increase the content of exchangeable potassium to at least 200 (mg K)/kg soil before the conventional application was performed, potassium chloride fertilizer equivalent to 274 (mg K)/kg soil was applied to the paddy fields before the cultivation trials.

In this study, it is assumed that the radiocesium that flowed into the two paddy fields from irrigation water, rainfall, fallout did not evaporate or permeate at site *K*. Therefore, we performed the evaluation according to the water balance between inflow through rainfall and irrigation water and outflow through drainage water. We cultivated paddy rice to quantify the behavior and balance of radiocesium in the paddy fields [both approximately 0.29 hectare (100 m × 29 m)]. The first paddy field, located in the southern part of the site, was irrigated with water from a pond; hereafter, this paddy field is denoted as K_S and its irrigation water as K_P. The other paddy field, located in the northern

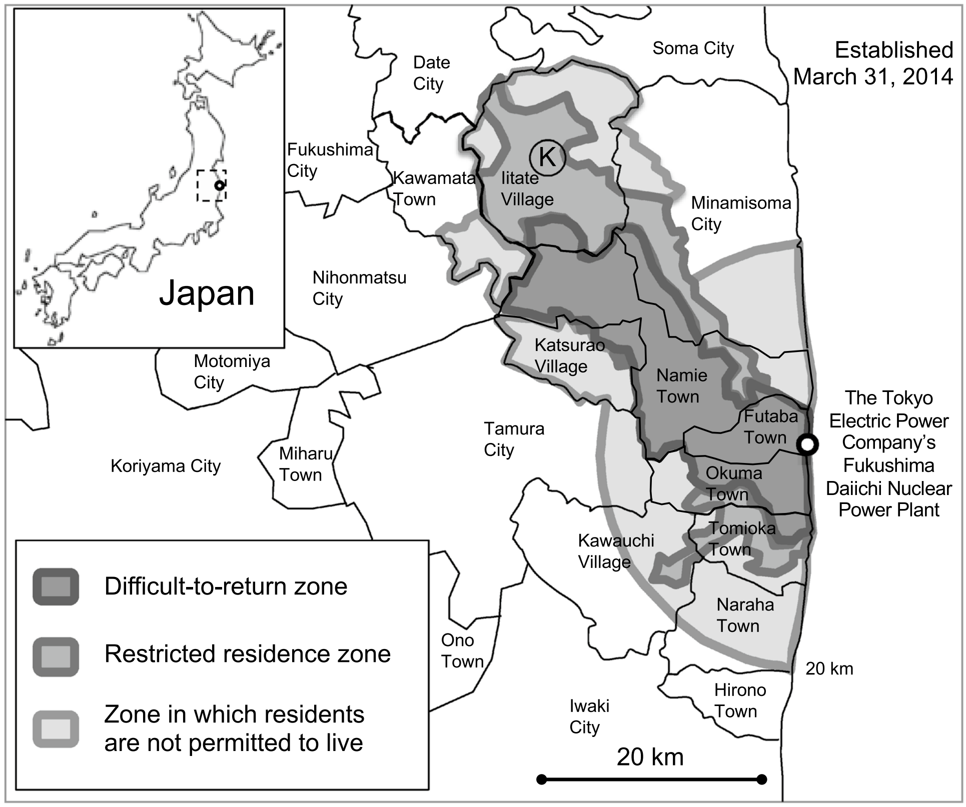

Fig. 1 Sampling sites used in the present study

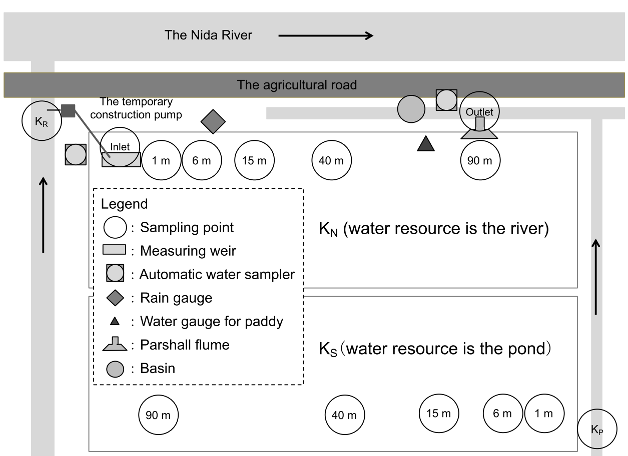

Fig. 2 Water sampling points and irrigation equipment at site *K*

part of the site, received river water that pumped through a measuring weir (220 cm × 40 cm × 40 cm) using a pump (KR-80; Koshin Co. Ltd., Kyoto, Japan). The inner diameter of the pump tube was 76.2 mm and the maximum flow rate was 950-L/min; hereafter, this paddy field is denoted as K_N and its irrigation water as K_R (Fig. 2).

The soil in site *K* is classified as a Gray Lowland soil (NIAES 2001). The Gray Lowland soil occupies approximately half of the paddy fields in Fukushima Prefecture (Cultivated Soil Classification Committee 1995). Some of the river's tributaries upstream of K_N contain ponds; therefore, water from these ponds might have affected our results. In addition, the water irrigating K_S passes through a farm ditch approximately 550 m long between the pond and the paddy. Consequently, the irrigation water might be affected by the soil at the bottom of the farm ditch.

Survey methods

To assess temporal changes in the concentrations of the different forms of radiocesium in the irrigation water, we obtained irrigation water samples 2–4 times per month during the sampling period in 2014. Water was sampled using buckets, plastic containers, pumps, and hoses. To analyze the water balance and the concentration of radiocesium in the water samples (irrigation, ponded, and drainage) in the paddy field, we installed an automatic water sampler (3700; Teledyne ISCO Co. Ltd., Nebraska, USA) and a water gauge (S&DLmini; OYO Corporation, Tokyo, Japan) at the inlet of site K_N to obtain 1-L samples every 10 min when irrigation occurred. We installed an automatic water sampler, a Parshall flume flow meter produced according to Japanese Industrial Standard (JIS) B7553 (2-inch type; Uizin, Tokyo, Japan), and a water gauge at the water outlet at site K_N to sample 1-L samples every 20 min when drainage occurred. We installed a rain gauge to obtain rainfall at site *K*. We did not install equipment to measure the amount of irrigation water at site K_S.

The irrigation volume from the inlet at site K_N was calculated with reference to the performance curve of the pump (discharge rate when the total pump head is 5 m = 900 L/min) and the height from the river to the inlet (Fig. 2). The irrigation time was the time during which the water level increased, according to the water gauge set on the measuring weir. The measurement was started after transplantation; therefore, the irrigation volume for puddling was not measured.

To analyze behavior and changes in the total (suspended plus dissolved) and dissolved radiocesium in the irrigation water flowing into the paddy fields, we measured the concentrations of radiocesium in the surface water of the paddy fields at sites K_N and K_S at various distances between the inlet and outlet. We blocked the outlet at a time when there was almost no surface water in the paddy field after ponding water was released in K_N (September 4) and K_S (August 21–22); we subsequently irrigated K_N and K_S at 2 h and 3 h, respectively. We obtained surface water samples from K_N and K_S at 5.5 h and 20 h, respectively, after the start of irrigation (Table 1). No rain fell during the sampling period of the surface water in the paddy fields. Sampling pumps and hoses with an inner diameter of 8 mm were carefully set up outside of the paddy fields to avoid disturbing the surface water and mixing the soil. We obtained 50-L samples for total and dissolved radiocesium (10-L × 1 and 20-L × 2) at a flow rate of 4 L/min at 1 m, 6 m, 15 m, 40 m, and 90 m from the inlet. (The sample obtained at x m is termed $K_N - x$, $K_S - x$, respectively; Fig. 2).

To estimate the impact of radiocesium fallout from rain and dust, we installed a stainless steel basin with a surface area of 0.5 m^2 at site K. We collected samples once or twice per month during the sampling period, using a silicon spatula and a plastic container. We filled the stainless steel basin with distilled water up to the 5-cm level to trap the air dust after installation and sampling.

We obtained soil and brown rice samples from near the inlet, at the center of the paddy, and near the outlet (indicated, for example, as $K_N - \mathrm{I}$, $K_N - \mathrm{C}$, and $K_N - \mathrm{O}$, respectively) at approximately 5–6 m from the farm ditch by using quadrat sampling on 3 Oct. In site K_S, the outlet was close to the inlet; therefore, the $K_S - \mathrm{O}$ sample was obtained at a distance of approximately 100 m from the inlet. The inlet, outlet, and sampling sites are shown in Fig. 3.

The rice samples were taken from 50 hills from each quadrat, which consisted of 3–4 rows. Soil samples were obtained from the plow layer (the upper 15 cm of soil) using a soil sample collector with 5 cm radius. We combined soil samples taken from four places within each quadrat.

Table 1 Water sampling time points and suspended solids (SS) values at site K in 2014

Irrigation time	K_N^a		K_S^a	
	9/4/2014 08:00–10:00		8/21/2014 15:00–18:00	
	Sampling time	SS (mg/L)	Sampling time	SS (mg/L)
Inlet	9/4 08:00–10:00	2.5	8/21 18:30	11.4
1 m	9/4 13:30	5.4	8/22 11:00	3.1
6 m	9/4 13:50	15.2	8/22 11:40	3.7
15 m	9/4 14:30	5.4	8/22 13:00	14.1
40 m	9/4 15:00	5.1	8/22 13:20	9.2
90 m	9/4 15:30	5.5	8/22 13:40	7.9

[a] K_N and K_S are paddy fields in the northern and southern parts of site K, respectively

Radiocesium analyses

We defined the total radiocesium as the concentration in the raw water of the sample, and the dissolved radiocesium as the concentration in the liquid passed through a membrane filter with 0.45-μm pores. We collected the water samples in 0.5-L and 2-L polypropylene plastic containers and 10-L and 20-L low-density polyethylene plastic containers. We transported these containers to the laboratory and performed the pre-processing procedures shown in Fig. 4. We separated the water samples into three fractions of total radiocesium (2–10 L), dissolved radiocesium (20–40 L), and suspended solids (0.5 L). We analyzed ^{134}Cs and ^{137}Cs in all the samples.

To analyze total radiocesium, we used the evaporative concentration method in a draft chamber to gradually reduce the sample volume from 10 L to approximately 2 L by using 2-L beakers and a hot plate. The radiocesium concentration

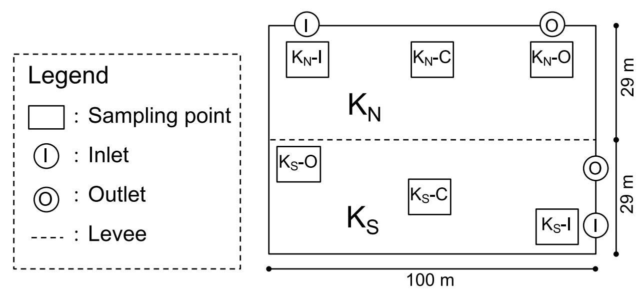

Fig. 3 Soil and brown rice quadrat sampling points

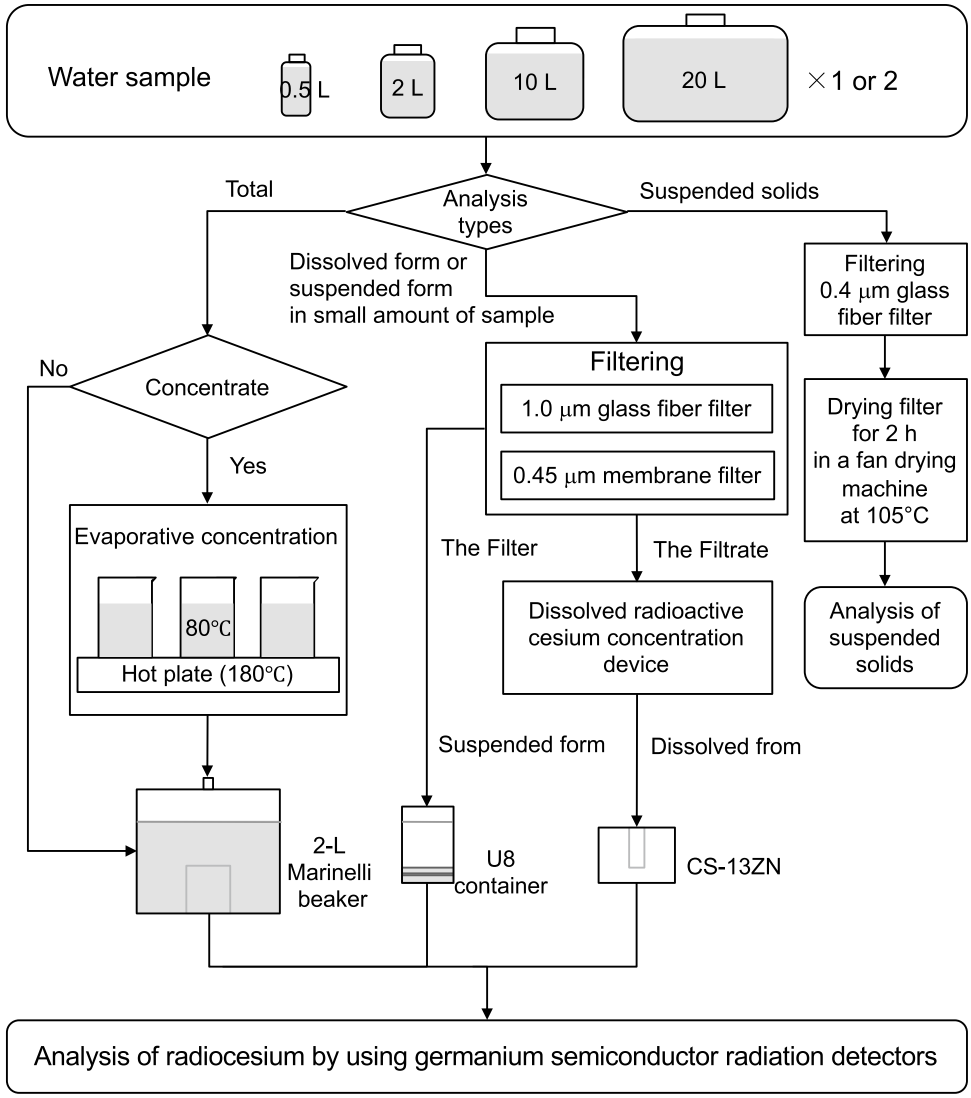

Fig. 4 Procedures for laboratory analysis

in the sample was measured using a 2-L Marinelli beaker. We analyzed the concentration for fallout samples in the stainless steel basin after pre-processing them in the same way as the samples used to analyze the total radiocesium concentration.

To process solid–liquid separation, we filtered the 20–40-L samples by using a glass fiber filter with a 150 mm diameter and pore diameter of 1.0 µm (GA-100; Advantec Co. Ltd., Tokyo, Japan) and a membrane filter with a 142 mm diameter and pore diameter of 0.45 µm (A045A; Advantec Co. Ltd.). Samples for which the volume collected by the automatic water sampler was small (approximately 2 L) were filtered using a 47-mm-diameter filter. The 1.0-µm and 0.45-µm filters of 47 mm diameter were used to measure the suspended radiocesium concentration.

To analyze dissolved radiocesium of the samples, we reduced the pre-processing and measurement time by adapting a method that used a nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (CS-13ZN; Japan Vilene Co. Ltd., Tokyo, Japan), developed by Yasutaka et al. (2015). We used a dissolved radiocesium concentration measurement device (AFC-550; Fujiwara Scientific Co. Ltd., Tokyo, Japan) developed by the National Institute of Advanced Industrial Science and Technology (Tsuji et al. 2014). The dissolved radiocesium was collected on the fabric within the cartridge. We used a nondestructive method (ICTRM 2015; Yasutaka et al. 2015) to calculate the concentration of dissolved radiocesium (Bq/L).

The radiocesium concentrations of the water, brown rice, and soil samples were determined with high-purity germanium (HPGe) detectors with relative efficiencies of 25% and 40%, respectively (GC4020-7500SL or GC2520-7500SL; Canberra Japan Co. Ltd., Tokyo, Japan). For all the samples, we measured to an accuracy of 10% or less of the relative standard deviation and applied attenuation compensation on November 1, 2014, for comparison.

Analyses of suspended solids and exchangeable potassium

To analyze the concentration of suspended solids, we filtered 0.5-L samples through a glass fiber filter with a pore size of 0.4 µm (Fig. 4). We selected brown rice samples by using a 1.8-mm mesh. Following radiocesium measurements, we applied a 15% moisture correction using the method for calculating brown rice weights in Japanese rice production guidelines (MAFF 2016). Soil samples were ground after natural drying and passed through a 2-mm mesh sieve. After radiocesium measurement, we corrected the concentration on an oven-dry basis (105 °C).

We measured the content of exchangeable potassium in the soil at the quadrat sampling locations at each site after cultivation trials. The exchangeable potassium was extracted using 1 M ammonium acetate (soil: solution = 1:10, 1-h shaking) at the room temperature, and potassium concentrations in the solution were determined using an atomic absorption spectrophotometer (ZA3000; Hitachi High-Technologies Corporation, Tokyo, Japan).

Calculation of the transfer factor (TF)

The TF of radiocesium from soil to brown rice varies depending on the soil, fertilizer, and other conditions (Fujimura et al. 2016b). We calculated the TF of radiocesium from soil to brown rice as follows (Eq. 1).

$$\mathrm{TF} = \frac{\text{Radiocesium concentration in brown rice (Bq/kg)}}{\text{Radiocesium concentration in dried soil (Bq/kg)}}, \tag{1}$$

where the brown rice has a 15% moisture correction after radiocesium measurement.

Consideration of the solid/liquid distribution coefficient

Radionuclides exist in a suspended or dissolved form in fresh water. The distribution of the dissolved and suspended forms of radiocesium is expressed by the solid/liquid distribution coefficient, Kd (IAEA 2010). The Kd of radiocesium in suspended environmental water is defined as the ratio of the radiocesium concentration in suspended solid form to the dissolved radiocesium concentration (JAERI 1995; Yamaguchi et al. 2012). In this study, the Kd for irrigation water was calculated using (Eq. 2), which modifies the suspended radiocesium based on the equation from Yamaguchi et al. (2012).

$$Kd = \frac{(T - D)/\mathrm{SS}}{D}, \tag{2}$$

where T is the total radiocesium concentration (Bq/L), D is the dissolved radiocesium concentration (Bq/L), and SS is the suspended solids concentration (kg/L).

Results

Temporal changes in the radiocesium concentration in irrigation and drainage water

The total and dissolved radiocesium concentrations at K_N were lower in drainage water than in irrigation water (Fig. 5). The average ratios (dissolved/total) for the radiocesium concentrations of the two irrigation water samples were 40.3% for K_N (K_R and $K_N - \mathrm{I}$ ratio) and 13.6% for K_S (K_P ratio) (Figs. 5, 6).

Spatial behavior and form changes of radiocesium in irrigation water

The total and dissolved radiocesium concentrations in the surface water in the both paddy fields, measured in the manner of Table 1 and Fig. 2, tended to decrease with increasing distance from the inlet (Fig. 7). However, at some sampling points located far from the inlet, the total concentrations of radiocesium were higher than those at sampling points located closer to the inlet (e.g., $K_S - 15$ compared with $K_S - 6$) (Fig. 7).

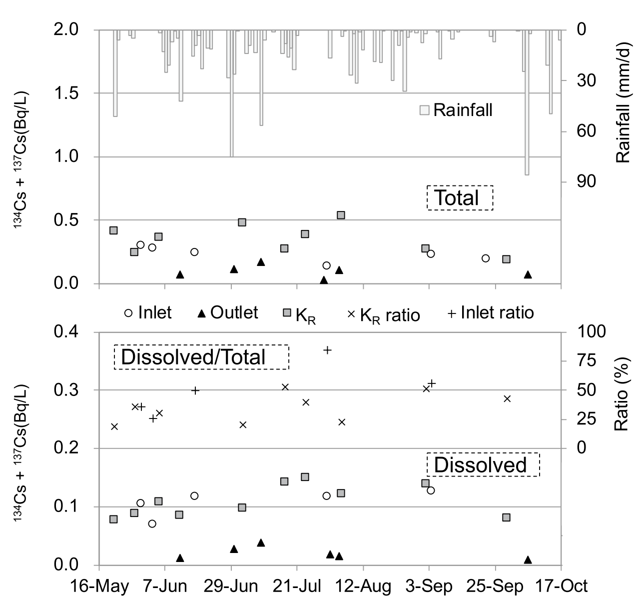

Fig. 5 Concentrations of radiocesium in the K_R (the irrigation water from the river at site K) inlet and outlet at site K_N (the northern part of site K) during the irrigation period in 2014

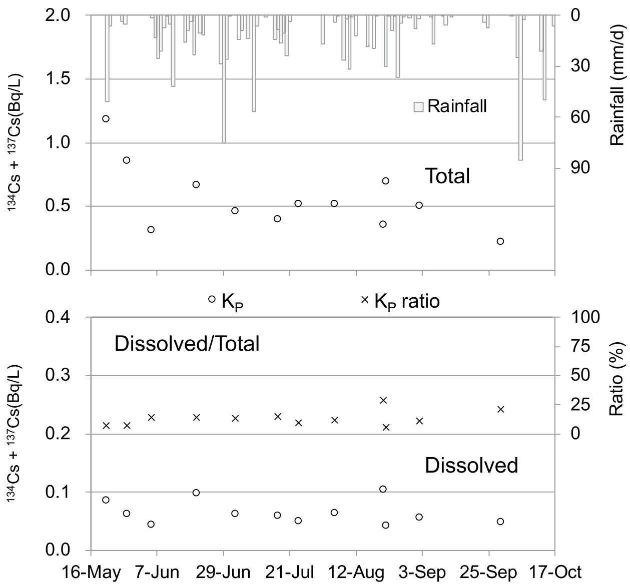

Fig. 6 Concentrations of radiocesium in the K_P (the irrigation water from the pond at site *K*) at site K_S (the southern part of site *K*) during the irrigation period in 2014

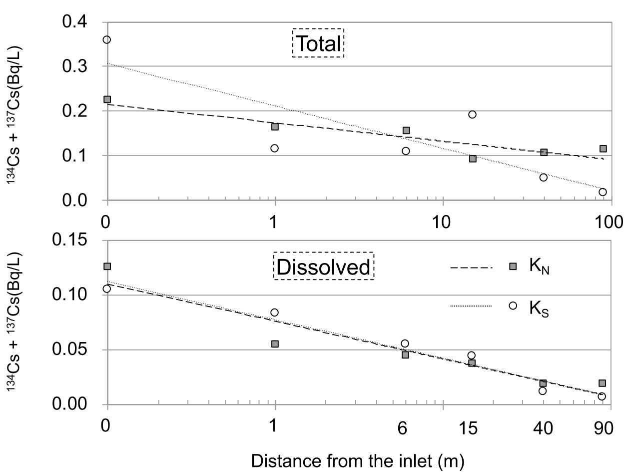

Fig. 7 Concentrations of radiocesium in the irrigation water in 2014 at various distances from the water inlet at sites K_N and K_S

Radiocesium concentration in fallout, soil, and brown rice

The time-weighted average fallout density during the entire sampling period was 0.125 Bq/(m^2 day), and the total deposit was 17.8 Bq/m^2. The time-weighted average ^{134}Cs/^{137}Cs ratio was 0.30 in atmospheric depositions (Table 2).

The content of exchangeable potassium (mg K)/kg in the soil and the concentration of radiocesium in the soil and brown rice are shown in Table 3. The soil to brown rice transfer factor (TF) at each sampling points was 0.0015–0.0068.

Quantification limits of ^{134}Cs and ^{137}Cs

Our measurements were to an accuracy of 10% or less of the relative standard deviation. The quantification limits of ^{134}Cs were 0.0013–0.25 Bq/L, 0.00080–0.043 Bq/(m^2 day), 0.16–1.05 Bq/kg, and 13.45–86.68 Bq/kg for water, fallout, brown rice, and soil samples, respectively. Similarly, the quantification limits of ^{137}Cs were 0.0013–0.17 Bq/L, 0.00092–0.052 Bq/(m^2 day), 0.17–1.14 Bq/kg, and 11.19–66.19 Bq/kg for water, fallout, brown rice, and soil samples, respectively.

Quantification of radiocesium in the irrigation and drainage water

From May 20 to October 17, 2014, the irrigation volume from the inlet at site K_N was 362 mm, and the inflow volumes of total and dissolved radiocesium were 95.3 Bq/m^2 and 37.2 Bq/m^2, respectively (Table 4). During the same period, the drainage volume from the outlet was 150 mm, and the outflow volumes of total and dissolved radiocesium were 14.7 Bq/m^2 and 3.2 Bq/m^2, respectively (Table 4). The rainfall at site *K* from May 16 to October 17 was 1010 mm, and the flux of water that flowed into K_N during this period was approximately 1372 mm. The surface water level at K_N from August 13 to 16 drying in the survey period was decreased 7.7 mm/day.

Ratio of solid/liquid radiocesium based on water source

The Kd ranged from 1.7 to 9.1 and 2.0 to 12.1 (10^5 L/kg) at K_N and K_S, respectively (Table 5). Furthermore, the average Kd value was approximately 1.3 times larger for K_S than for K_N.

Discussion

Quantification of the radiocesium balance in the paddy fields

Shin et al. (2015) reported that the amount of radiocesium inflow in irrigation water for a decontaminated paddy field was 0.031% of the amount deposited by scattering of radiocesium at the time of the FDNPP accident in difficult-to-return zone from July to October 2013. However, the behavior of radiocesium in brown rice, fallout, and soil in decontaminated paddy fields remains unclear. Miyazu et al. (2016) reported that total outflow of ^{137}Cs from the fields

Table 2 Inflow volume of radiocesium in atmospheric depositions at site *K* in 2014

Sampling period	Rainfall (mm)	^{134}Cs Bq/(m^2 day)	^{137}Cs Bq/(m^2 day)	^{134}Cs + ^{137}Cs Bq/(m^2 day)	^{134}Cs/^{137}Cs
5/20–6/12 (23 days)	195.5	0.001	0.003	0.004	0.26
6/12–6/30 (18 days)	210.5	0.005	0.023	0.028	0.21
6/30–7/17 (17 days)	131.5	0.007	0.011	0.018	0.62
7/17–8/1 (15 days)	66.0	0.023	0.054	0.076	0.42
8/1–8/21 (20 days)	124.0	0.167	0.588	0.755	0.28
8/21–9/4 (14 days)	93.0	0.003	0.013	0.017	0.25
9/4–10/9 (35 days)	149.0	0.004	0.010	0.014	0.41
Time-weighted average		0.029	0.097	0.125	0.30
Total Bq/(m^2 142 days)[a]	969.5	4.070	13.718	17.787	0.30

[a]The unit is only the total volume of radiocesium during sampling period

Table 3 The content of exchangeable potassium in the soil, concentrations of radiocesium (Bq/kg) in the soil and brown rice, and transfer factor (*TF*) of radiocesium from soil to brown rice after cultivation trials in 2014

	K_N				K_S^a			
	Potassium (mg K)/kg	Soil (Bq/kg)	Rice (Bq/kg)	Transfer factor (TF)	Potassium (mg K)/kg	Soil (Bq/kg)	Rice (Bq/kg)	Transfer factor (TF)
Inlet	560	810	1.64	0.0020	245	377	1.75	0.0046
Center	449	1087	1.65	0.0015	240	390	2.67	0.0068
Outlet	415	898	1.32	0.0015	322	501	1.51	0.0030
Average	475	932	1.54	0.0017	269	423	1.98	0.0047

[a]The K_S-O sample was obtained approximately 100 m from the inlet of K_S, the paddy field in the southern part of site *K*, not near the outlet (Fig. 3)

Table 4 Inflow and outflow volumes of radiocesium at site K_N (2900 m^2), in 2014

Period	Irrigation volume (mm)	Inflow volume of radiocesium[a] (Bq/m^2)		Period	Drainage volume (mm)	Outflow volume of radiocesium[a] (Bq/m^2)	
	Inlet (a)	Total (a) × (Bq/L)	Dissolved (a) × (Bq/L)		Outlet (a)	Total (a) × (Bq/L)	Dissolved (a) × (Bq/L)
5/20–6/1	21.7	6.16	1.89	5/20–6/11	28.3	1.97	0.33
6/2–6/6	57.4	15.68	3.99	6/12–6/29	59.7	5.40	1.17
6/7–6/11	48.1	14.32	3.71	6/30	16.2	1.80	0.44
6/12–6/19	37.2	9.50	3.76	7/1–7/8	3.5	0.49	0.11
6/20–7/10	10.9	3.85	1.16	7/9	18.8	3.17	0.72
7/11–7/29	125.7	32.58	16.70	7/10–7/29	7.1	0.70	0.20
7/30–9/4	34.1	7.72	4.30	7/30–8/4	11.9	0.81	0.21
9/5–10/17	26.4	5.53	1.66	8/5–10/17	4.5	0.40	0.03
Total	362	95.3	37.2	Total	150	14.7	3.2

[a]The volume of radiocesium was estimated as the volume using the water balance (excluding puddle water) to the paddy field via irrigation or drainage water

was 0.003–0.028% during puddling and 0.001–0.011% during midsummer drainage of already existing ^{137}Cs in the paddy field.

At site K_N, the total annual balance of total radiocesium and dissolved radiocesium in irrigation water was estimated to be 80.6 Bq/(m^2 sampling period) and 34.0 Bq/(m^2 sampling period), respectively (Table 6). The outflow of total radiocesium [drainage water: 14.7 Bq/(m^2 sampling period)] was 13.0% of the inflow to the paddy field [irrigation water: 95.3 Bq/(m^2 sampling period) plus fallout: 17.8 Bq/(m^2 sampling period)] at site K_N (Table 6).

Table 5 Suspended solids (*SS*) and solid/liquid distribution coefficient (Kd) values for various sampling time points in 2014

Sampling date	K_N		K_S	
	SS (mg/L)	Kd (10^5 L/kg)	SS (mg/L)	Kd (10^5 L/kg)
5/21	9.4	4.6	65.1	2.0
5/28	5.7	3.1	24.2	5.3
6/5	5.6	4.2	4.9	12.1
7/3	4.3	9.1	24.8	2.5
7/17	4.7	1.9	15.9	3.6
7/24	7.1	2.2	19.7	4.6
8/5	6.1	5.5	17.7	4.0
9/2	5.6	1.7	11.3	7.1
9/29	2.3	5.8	4.5	8.0
Average	5.6	4.2	20.9	5.5

The values were calculated using the data for irrigation water

The balance of total radiocesium [98.4 Bq/(m^2 sampling period)] was 0.076% of the already existing radiocesium in the decontaminated paddy field, located ca. 40 km from FDNPP [if apparent specific gravity (dry bulk density) of a Gray Lowland soil was 0.93 Mg/m^3 of measured value of K_N. The amount of the plow layer (the upper 15 cm of soil) was 139.5 kg/m^2, and the amount of radiocesium in the K_N plow layer was 130 kBq/m^2 (932 Bq/kg × 139.5 kg/m^2)]. At all sampling points in site *K*, the soil and brown rice transfer factor (TF) value ranged from 0.0015 to 0.0068 and was within levels recorded before the FDNPP accident (0.00021 to 0.012) by Tsukada et al. (2002). Gravitational irrigation could possibly increase the flux of inflow of the pump irrigation at K_N. As the amount of irrigation increases, it could affect concentration of radiocesium in brown rice. If the ratio of discharge to amount of irrigation assumed to be constant, the balances were estimated at 111–311 Bq/(m^2 sampling period) at an irrigation volume of 500–2000 mm (Table 6).

Behavior of radiocesium in the surface water of paddy fields

The average Kd value was approximately 1.3 times larger for K_S than for K_N (Table 5). We believed that radiocesium in the irrigation water from ponds was more likely to be adsorbed to the solid phase than radiocesium from rivers. However, because the difference in Kd was small, further studies are required to completely elucidate the behavior of the transfer of radiocesium from irrigation water sources of multiple sites.

The amount of dissolved radiocesium, which can be absorbed by crops, declined by 60% and 80% within 15 m and 40 m of the inlet, respectively, and total radiocesium also tended to decrease toward outlet (Fig. 7). This finding indicates that the dissolved radiocesium was either directly absorbed by rice, algae, and other plants, or it was adsorbed onto soil particles, and the suspended

Table 6 Estimated volume of radiocesium balance at site K_N in 2014

Site	K_N					
Area (m^2)	2900					
Sampling period	5/20–10/17	5/20–10/9		5/20–10/17		
Type	Irrigation water	Fallout	Total	Irrigation water		
Irrigation volume (mm)	(b) 362			(f) 500	(g) 1000	(h) 2000
Drainage volume (mm)	(c) 150			(c) ÷ (b) × (f) = (i) 207	(c) ÷ (b) × (g) = (j) 414	(c) ÷ (b) × (g) = (k) 829
(1) Average	(Bq/L)	(Bq/m^2 day)				
Total	0.288	0.125				
(Dissolved)	(0.108)					
(2) Inflow [Bq/(m^2·period)]	(d)			(d) ÷ (b) × (f)	(d) ÷ (b) × (g)	(d) ÷ (b) × (h)
Total	**95.3**[a]	**17.8**[a]	113.1	131.6	263.2	516.5
(Dissolved)	(**37.2**[a])			(51.4)	(102.8)	(205.5)
(3) Outflow [Bq/(m^2 period)]	(e)			(e) ÷ (c) × (i)	(e) ÷ (c) × (j)	(e) ÷ (c) × (k)
Total	**14.7**[a]		14.7	20.3	40.6	81.1
(Dissolved)	(**3.2**[a])			(4.4)	(8.8)	(17.7)
Balance [Bq/(m^2 period)]						
(2)–(3) Total	80.6	17.8	98.4	111.3[b]	222.6[b]	311.0[b]
(Dissolved)	(34.0)			(47.0)	(94.0)	(187.8)

[a]The bolded values are estimated the radiocesium amounts indicated in Tables 2 and 4

[b]The balances were estimated for the irrigation volumes of (f), (g), and (h)

substances including radiocesium in irrigation water sedimented in the paddy field during nonrainfall.

Radionuclides in soil–water environments migrate vertically in solution, as colloids with infiltration water flow, or attached to fine soil particles. Transport of radiocesium in solution by infiltration is slower than the water flow because of sorption–desorption and fixation on soil particles (Konoplev et al. 2016). Considering the short time span (5.5–22.7 h) between irrigation and sampling and assuming the radiocesium concentration in irrigation water and the amount of adsorption to the soil (IAEA 2010) are in equilibrium, the reduction of the concentration of dissolved radiocesium was probably caused by adsorption to the soil with decreasing water flow after flowing into paddy fields with a lot of soil particles. However, it is necessary to evaluate the adsorption to soil particles by residence time of surface water and the contact time settled on the bottom of the paddy in future studies.

Origin of radiocesium

After the FDNPP accident, most of the radiocesium fallout was retained in the soil. The Kd value in the area was 10^{4-6} L/kg (Yoshimura et al. 2015; Tsukada et al. 2017), and the Kd measured in this study was 1.7×10^5–1.2×10^6 L/kg. Dry and windy weather conditions were expected to result in further contamination, as wind-blown particles of soil containing radiocesium were scattered to new locations (Watanabe 2014).

The inflow volume of radiocesium by atmospheric deposition was 0.755 Bq/(m^2 day) on August 1–21, 2014 (Table 2). This value was higher than that during the other investigated periods. In August 2014, typhoon 11 hit Japan, blowing strong winds across the country (JMA 2014) and possibly causing further fallout.

The average radiocesium fallout density (including rainfall) from rain and dust during the entire sampling period was 0.125 Bq/(m^2 day), and the total deposit was 17.8 Bq/(m^2 sampling period) (Table 2). Therefore, the effect of the atmospheric fallout on paddy soil was expected to be negligible, because the volume was 0.012% of the amount of radiocesium in the decontaminated paddy field at K_N. It is possible that radiocesium from the fallout directly adhered to rice plants and was taken up by the brown rice. However, the effect of the atmospheric fallout on rice at the sampled site was limited in this study.

The radiocesium of the sampling sites was thought to be derived from the accident, considering the half-life (^{134}Cs: 2.1 year, ^{137}Cs: 30.2 year) of radiocesium, because ^{134}Cs was detected in all samples in the present study.

Impact on the radiocesium concentration in brown rice

In contrast to the concentration of radiocesium in the surface water in the paddy field, the concentration in the soil was different despite being adjacent between the investigated K_N and K_S (Table 3). This variability might be attributed to the difference in removal efficiency of radiocesium during the official decontamination program (MAFF 2013b). On the other hand, the radiocesium concentration in the brown rice did not differ much and it was low. These low concentrations were much lower than the standard limit in Japan (100 Bq/kg) (MHLW 2012).

The content of exchangeable potassium was > 200 (mg K)/kg after cultivation trials (Table 3), potassium fertilization was suspected of suppressing radiocesium uptake in brown rice due to the antagonism between potassium and radiocesium, and this information, together with the low variability between samples, suggests that the radiocesium concentration in the soil and irrigation water had little effect on the radiocesium concentration in brown rice when there was enough potassium in paddy soil.

Conclusions

In the present study, we quantitatively analyzed the balance of radiocesium and monitored its behavior at two decontaminated paddy fields located ca. 40-km northwest of FDNPP in 2014.

Our study yielded several key findings. First, at the investigated site K at the 40-km zone, the total and dissolved concentrations of radiocesium flowing into a decontaminated paddy field via irrigation water were at least 95 Bq/(m^2 sampling period) and 37 Bq/(m^2 sampling period), respectively, in 2014.

Second, about 85% of the radiocesium in the irrigation water did not drain away but accumulated in the paddy field. However, in comparison with the amount of the radiocesium deposited in the fields, the amount of radiocesium that accumulated due to irrigation was approximately 0.076%. The outflow of total radiocesium from the paddy field was 13.0% of that in the inflow to the paddy field (irrigation water and fallout) at site K_N.

Third, the concentration of total and dissolved radiocesium in the irrigation water tended to decline rapidly with the increasing distance from the inlet in the paddy fields. Moreover, accumulation was higher near the inlets than at the outlets. Radiocesium that settled in the paddy field was likely adsorbed onto soil particles.

Fourth, the soil to brown rice transfer factor (TF) was from 0.0015 to 0.0068 in the decontaminated paddy fields in which soil improvement was conducted to increase the content of exchangeable potassium to 200 (mg K)/kg soil before the conventional application in 2014.

Finally, at all the sampling point, the concentration of radiocesium in brown rice was about 2% of the standard limit in Japan (100 Bq/kg), and the impact on brown rice from radiocesium in the irrigation water was limited.

It may be possible for farming to recommence in regions where decontamination of agricultural land has been completed, but continuous monitoring will be required.

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