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Fate of radiocesium in freshwater aquatic plants and algae in the vicinity of the Fukushima Daiichi nuclear power plant

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Abstract The behavior of radiocesium (^{137}Cs) in aquatic plants (five species) and algae (three genera) grown in either a river (one sampling point) or pond (four sampling points) in the vicinity of the Fukushima Daiichi nuclear power plant was investigated. The ¹³⁷Cs concentration of <0.45-µm fractions of water taken from the river and ponds was between 5.01 \times 10⁻¹ and 2.98 Bq/L, while that of sediment was between 4.85×10^3 and 5.72×10^4 Bg/kg dry weight. The ratio of ¹³⁷Cs concentration of sediment/ water in ponds was $\sim 10^4$. The sediment-to-plant transfer factor (TF) [(137 Cs concentration Bq/kg dry weight_{plant}) × $(^{137}Cs \text{ concentration Bq/kg dry weight})^{-1}]$ was also measured. For aquatic plants, the highest value was 5.55 for Potamogeton crispus from the river, while the lowest was 3.34×10^{-2} for *P. distinctus* from a pond. There were significant differences in values between aquatic plants belonging to the same genus. The water-to-plant TF [(¹³⁷Cs concentration Bq/kg dry weight_{plant}) × (137 Cs concentration Bq/ L_{water})⁻¹] of filamentous algae (*Spirogyra* sp.) and cyanobacteria (coexisting Anabaena sp. and Microcystis sp.) were 2.39×10^3 and 1.26×10^3 , respectively. The ¹³⁷Cs concentration of cyanobacteria in pond water was 4.87×10^{-1} Bq/L, which was the same order of magnitude as the ¹³⁷Cs concentration of pond water. Enrichment of ¹³⁷Cs in cvanobacteria was not observed.

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⊠ Yoshito Sasaki sasaki.yoshito@jaea.go.jp **Keywords** Radiocesium · Fukushima Daiichi nuclear power plant · Aquatic plants · Algae

Introduction

As a result of the accident at Fukushima Daiichi nuclear power plant (FDNPP) caused by the Great East Japan earthquake on 11 March 2011, a large amount of radionuclides—such as cesium (134 Cs, 2.06-year half-life; 137 Cs, 30.2-year half-life) and iodine (131 I, 8.04-day half-life) were released into the environment (Chino et al. 2011). Of major concern is the presence of radiocesium (137 Cs), which has a long half-life and thus has a continuing presence in the environment. However, the long-term nuclide migration of 137 Cs in the environment is not well understood.

Radiocesium has been detected in fish inhabiting rivers in the Fukushima Prefecture (Ministry of the Environment 2013, 2014), which becomes incorporated in their bodies because of biological and physical cycles (Avery 1996) involving aquatic plants and algae in freshwater. Polar and Bayülgen (1991) reported that aquatic plants concentrate ¹³⁷Cs more than terrestrial plants. Fukuda et al. (2014) reported that several species of aquatic plants and algae had high radionuclide recovery efficiency under culture conditions in the absence of potassium but in the presence of ¹³⁷Cs.

There have been many studies regarding ¹³⁷Cs transfer in terrestrial plants but few reports on aquatic plants (Avery 1996). Although one report exists regarding the transfer of ¹³⁷Cs released from the FDNPP to algae in seawater (Kawai et al. 2014), thus far there have been no studies regarding this transfer in freshwater. In this study, ¹³⁷Cs concentrations of water, sediment, aquatic plants, and algae in a river and artificial agriculture ponds within \sim 7 km of the FDNPP were examined. As well, transfer factors (TFs)—

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representing the ratio of incorporation of radionuclides from sediment and water into plants—were compared.

Materials and methods

Sampling

We investigated the transfer of ¹³⁷Cs to aquatic plants and algae in ponds and a river in the region contaminated with radionuclides released by the accident at the FDNPP. Locations of the river (one point: KUMR2) and ponds (four points: OKUMA24, OKUMA70, FUTABA55, and FUTABA48) from which samples were obtained are shown in Fig. 1. Samples were collected from July to September 2013 using a 225-cm² Ekman-Birge bottom sampler (RIGO, Tokyo, Japan) at the midpoint of the pond, and with a plastic ladle (4 L; shaft length, 120 cm) (SANKA, Nigata, Japan) for the river. Collected sediment samples were dried to a constant weight at 105 °C. After being mixed to ensure consistency, sediment samples were placed in 80-ml polystyrene containers (V7 containers) for measurement of ¹³⁷Cs concentration. Water depth measurements were obtained from the center of the ponds and at the aquatic plant collection points of the river. Measurements of pH in the ponds were obtained using a multiparameter waterquality-meter multimonitoring system (W-23XD, HOR-IBA, Ltd., Kyoto, Japan). In the river, a pH meter (B-712, HORIBA, Ltd.) was used after water sampling.

After their fresh weight was measured, collected aquatic plants and algae were dried to a constant weight at 90 °C in a forced convection constant temperature oven (DMK300, Yamato Scientific Co., Ltd., Tokyo, Japan). Cyanobacteria were collected in the vicinity of the pond intake, which is where they were concentrated. Surface water containing cyanobacteria that was sampled using a ladle was passed through a sieve (mesh size, 250 µm) to remove debris. Once the surface water was centrifuged (6654 g, 5 min, 3-16 L, Sigma Laborzentrifugen GmbH, Osterode am Harz, Germany), the algal precipitate was collected. After their fresh weight was measured, collected algal fractions were placed on a Durapore[®] polyvinylidene fluoride membrane filter (pore size, 0.45 µm; diameter, 47 mm; HVLP4700, Merck Millipore, MA, USA) and dried to a constant weight at 90 °C in the forced-convection constant-temperature oven (DMK300) before measuring ¹³⁷Cs concentration. The percent moisture content of aquatic plants and algae was determined using the following equation:

$$\begin{aligned} \text{Percent moisture content} &= (\text{Fresh weight} - \text{Dry weight}) / \\ \text{Fresh weight} \end{aligned} (1)$$

Fig. 1 Sampling points and Fukushima Daiichi Nuclear Power Plant ► (FDNPP). Squatic plants and algae obtained from the river and ponds around the FDNPP: **a** *Potamogeton crispus* L., **b** *Trapa bispinosa* Roxb. var. *japonica* Nakai, **c** *Nymphaea tetragona* Georgi, **d** *Potamogeton distinctus* A. Benn. **e** *Spirogyra* sp., **f** coexisting *Anabaena* sp. and *Microcystis* sp.

Identification of aquatic plants and algae

Aquatic plants were identified based on their morphology, with reference to Kadono (1994). Algae were also identified based on their morphology, which was observed using a microscope (VHX-2000, Keyence Corporation, Osaka, Japan), with reference to Hirose and Yamagishi (1977).

Measurement of ¹³⁷Cs concentration

Radiocesium concentrations of water (<0.45-µm fractions in 500-ml plastic containers), dried sediment [in 80-ml plastic (V7) containers], dried aquatic plants (in 100-ml containers), and cyanobacteria (on 47-mm-diameter membrane filters) were measured using an n-type, high-purity Ge-detector (GMX40P4-76 germanium detector, Seiko EG&G ORTEC, Tokyo, Japan) with 40 % relative efficiency. Gamma-ray emission at 604 keV (¹³⁴Cs) and 661 keV (137Cs) was also measured. For the pulse-height analysis, a multichannel analyzer (MCA7600, Seiko EG&G ORTEC) was used in line with spectrum analysis software (Gamma Studio, Seiko EG&G ORTEC). Efficiency calibration was carried out with a multiple gammaray-emitting standard source (including ten nuclides) packed in the same type of vessel (Eckert and Ziegler Isotope Products, CA, USA) or in a plastic disc with the same active area (ϕ 42 mm) as the membrane filter (Eckert and Ziegler Nuclitec GmbH, Braunschweing, Germany). Radiocesium decay was corrected on the sampling dates. The 137 Cs concentration measurements showed a 1σ counting error.

Calculation of TF

Sediment-to-plant and water-to-plant TFs were determined using Eqs. (2) and (3), where C^p is the plant ¹³⁷Cs concentration (Bq/kg dry weight), C^w is the water ¹³⁷Cs concentration (Bq/L), and C^s is the sediment ¹³⁷Cs concentration (Bq/kg).

$$\mathbf{C}^{\mathbf{p}} = \text{Water-to-plant TF} \times \mathbf{C}^{\mathbf{w}}$$
(2)

$$C^{p} =$$
Sediment-to-plant $TF \times C^{s}$ (3)



50 km



Results and discussion

Pond FUTABA48, in which blue-green algae (cyanobacteria) was found, was 240-cm deep, which was deeper than ponds containing aquatic plants rooted to the bottom (depth, 120–170 cm). The pH of pond water increases due to consumption of carbon dioxide through photosynthesis associated with the blooming of cyanobacteria (Paerl and Ustach 1982); therefore, the pH of pond FUTABA48 was higher; and that of water near the intake, where the cyanobacteria were concentrated, was 9.1 and was even higher in the center of the pond. The metabolism of cyanobacteria is influenced by changes in pH (Coleman and Colman 1981, Wang et al. 2011), whereas the effect of pH changes on ¹³⁷Cs concentration in cyanobacteria is unknown. With the exception of pond FUTABA48, the pH of surface water in the ponds and river was ~7 (Table 1).

The ¹³⁷Cs concentration in surface water and sediment is listed in Table 1. The highest concentration of ¹³⁷Cs in surface water was 2.98 Bq/L in FUTABA55, while the lowest was 5.01×10^{-1} Bq/L in KUMR2. The highest concentration in sediment was 5.72×10^4 Bq/kg in FUTABA55, while the lowest was 4.85×10^3 Bq/kg in KUMR2. After the Chernobyl nuclear power plant accident, concentration of ¹³⁷Cs in river water was estimated by catchment inventory of ¹³⁷Cs (Santschi et al. 1990, Smith et al. 2004, 2005). The same behavior of ¹³⁷Cs was observed in the Fukushima River after the FDNPP accident (Yoshimura et al. 2015). The ratio of ¹³⁷Cs concentration of sediment/water in the ponds was $\sim 10^4$ (Table 1), suggesting that ¹³⁷Cs concentration in water and sediment is in equilibrium.

Sakaguchi et al. (2015) reported that ¹³⁷Cs in the river water (<0.45 µm fraction) was present exclusively as the dissolved species rather than being adsorbed on suspended solids or complexed with organic materials. Dissolved ¹³⁷Cs species in the water are easily incorporated into plants. However, further investigation of the relationship between plant growth and dissolved ¹³⁷Cs concentration is required. Photographs of aquatic plants and algae from the river and ponds in the vicinity of the FDNPP are shown in Fig. 1. The moisture content and ¹³⁷Cs concentration of aquatic plants and algae are shown in Table 1. The water content of aquatic plants was ~ 90 %, with that of algae being higher. The ¹³⁷Cs concentrations of aquatic plants and algae were between 10^3 and 10^4 Bq/kg. The 137 Cs concentration of *Potamogeton crispus* was 2.69×10^4 Bg/ kg, which was the highest value for the collected aquatic plants. The ¹³⁷Cs concentration of Trapa bispinosa obtained from an adjacent pond, with similar water depth and pH, was similar, as the ¹³⁷Cs concentrations in the sediment and water of the pond were nearly identical. T. bispinosa seeds contain starch and are edible; however, seeds were not collected as this study was performed prior to fruiting. The ¹³⁷Cs concentrations of Nymphaea tetragona and P. distinctus were 10^3 Bq/kg, which was lower than the values for other aquatic plants. The cyanobacteria consisted of coexisting Anabaena sp. and Microcystis sp. The ¹³⁷Cs concentrations of filamentous algae and cyanobacteria were 10³ Bq/kg. Adsorbed ¹³⁷Cs on suspended solids of the clay and silt fraction is the main contributor to the transport of ¹³⁷Cs in water (Matsunaga et al. 2015). Cesium incorporated into clay minerals will not readily enter subsequent biological cycles as it is strongly adsorbed (Cremers et al. 1988; Valcke and Cremers 1994). In addition to the contribution from suspended solids, the movement of ¹³⁷Cs-contaminated microalgae with water flow leads to the potential spread of contamination. The ¹³⁷Cs concentration of the cyanobacterial fraction was $4.87 \times 10^{-1} \pm 1.74 \times 10^{-2}$ Bq/L, which was the same order of magnitude as the ¹³⁷Cs concentration in the water. In this case, ¹³⁷Cs migration associated with the movement of cyanobacteria is limited as the ¹³⁷Cs concentration of cyanobacteria was low. As there is no data regarding changes in cell number and/or species of cyanobacteria, further investigation is required.

Calculated TF values are given in Table 1. The highest sediment-to-plant TF in aquatic plants was 5.55 for *P. crispus*, while that of T. bispinosa was between 4.46×10^{-1} and 1.10. Although P. crispus and P. distinctus belong to the same genus, there was a difference of two orders of magnitude in their sediment-to-plant TF values. There is also a difference in the growth form of these two species, with P. crispus having submerged leaves and P. distinctus having floating leaves. The sediment-to-plant TF values of N. tetragona and P. dis*tinctus* were 8.30×10^{-2} and 3.34×10^{-2} , respectively, which were lower than those of *T. bispinosa*, though all three species have floating leaves. However, each aquatic plant grew under different conditions in the river and ponds, and the impacts of these differences in growth form and growing conditions on sediment-to-plant TF are unknown. As an example of the impact of these differences, TFs of a cultivated cabbage were 2.1×10^{-3} and 3.3×10^{-1} , differing by two orders of magnitude as a result of different cultivation conditions (Tsukada and Hasegawa 2002). To clarify these effects, further investigation is required.

Soil-to-plant TF values $[(^{137}Cs \text{ concentration Bq/kg dry weight}_{plant}) \times (^{137}Cs \text{ concentration Bq/kg dry weight}_{soil})^{-1}]$ of wild terrestrial plants grown in arable land contaminated by the FDNPP accident were between 6×10^{-3} and 7×10^{-1} (Yamashita et al. 2014), which were lower than the sediment-to-water TF values of *P. crispus* and *T. bispinosa*. This is potentially due to the fact that the aquatic plants were grown in conditions that allowed for easy incorporation of dissolved ¹³⁷Cs, compared with the terrestrial plants.

Investigation site	Sampling date	Distance from FDNPP (km)		Depth of water (cm)	pH ¹³⁷ Cs concentra surface water (tion of ¹³⁷ Cs concentration of Bq/L) sediment (Bq/kg)		Ratio of ¹³⁷ Cs concentration of sediment/water		
KUMR2	2013 4.5			80 7.3		5.01×10^{-1}		4.85×10^{3}		9.7×10^{3}	
	Aug. 28					$\pm 1.27 \times 10^{-1}$		$\pm 6.51 \times 10^{-10}$	$)^{2}$	± 2.8	$\times 10^3$
OKUMA24	2013 Jul 17	2.5		140	6.7	1.05		3.32×10^{4}		$3.2 \times$	10^{4}
						$\pm 1.98 \times 10^{-1}$		$\pm 2.15 \times 10^{\circ}$	$)^{2}$	± 6.0	$\times 10^3$
OKUMA70	2013 Jul	2.5		120	6.8	9.60×10^{-1}		1.78×10^{4}		$1.9 \times$	10^{4}
	18					$\pm 1.99 \times 10^{-1}$		$\pm 1.41 \times 10$	$)^{2}$	± 3.8	$\times 10^3$
FUTABA55	2013 Jul	7		170	7.1	2.98		5.72×10^{4}		$1.9 \times$	10^{4}
	25					$\pm 7.30 \times 10^{-1}$		$\pm 2.69 \times 10^{-10}$	$)^{2}$	±4.7	$\times 10^3$
FUTABA48	2013 Jul 26	5.5		290	8.6	8.00×10^{-1}		9.49×10^{3}		$1.2 \times$	10 ⁴
						$\pm 2.03 \times 10^{-1}$		$\pm 1.27 \times 10^{-10}$	0^{2}	± 3.0	$\times 10^3$
Investigation site	Common n of the taxo	Common name of the taxon		Scientific	name	% H ₂ O dry weight	¹³⁷ Cs cor of plant	ncentration (Bq/kg)	Water-to-p transfer fa	olant ctor	Sediment-to-plant transfer factor
KUMR2	Aquatic plant		2013 Aug. 28	Potamogeton crispus		90	2.69×10^{4}		$5.37 \times 10^4 \pm 1.36 \times 10^4$		5.55
							$\pm 3.29 \times 10^2$				$\pm 1.01 \times 10^{-1}$
OKUMA24	Aquatic plant		2013 Jul	Trapa bispinosa		91	1.48×1	0^4	1.41×10^{-1}	4	4.46×10^{-1}
			17				$\pm 2.52 \times 10^2$		$\pm 2.67 \times 10^{3}$		$\pm 8.12 \times 10^{-3}$
OKUMA70	Aquatic plant		2013 Jul	Trapa bispinosa		90	1.96×1	0^4	2.04×10^{-10}	4	1.1
			18				$\pm 2.30 \times 10^2$		$\pm 4.24 \times 10^3$		$\pm 1.56 \times 10^{-2}$
	Aquatic plant		2013 Jul	Nymphaea		89	4.75 × 1	0^{3}	1.59 × 10	3	8.30×10^{-2}
			18	tetragon	ıa		$\pm 9.66 \times$	10^{2}	$\pm 3.92 \times 1$	10^{2}	$\pm 1.73 \times 10^{-3}$
FUTABA55	Aquatic plant		2013 Jul	Potamogeton		93	1.91 × 1	0^{3}	6.41×10^{-10}	2	3.34×10^{-2}
			18	distinctı	lS		± 5.27 \times	10^{2}	$\pm 1.58 \times 1$	10^{2}	$\pm 9.35 \times 10^{-4}$
	Alga (Filamentous Alga)		2013 Jul	<i>Spirogyra</i> sp.		97	7.11 × 1	0^{3}	2.39×10^{-10}	3	1.24×10^{-1}
			25				$\pm 1.94 \times$	10 ²	$\pm 5.88 \times 1$	10^{2}	$\pm 3.44 \times 10^{-3}$
FUTABA48	Alga		2013 Sep	*Anabaena sp.,		99	1.01×1	0^{3}	1.26×10^{-1}	3	1.06×10^{-1}
	(Cyanoba	cteria)	2	Microcys	<i>stis</i> sp).	$\pm 3.61 \times$	10 ²	$\pm 3.24 \times 1$	10^{2}	$\pm 1.47 \times 10^{-2}$

Table 1 Investigation site and date, pH of water, radiocesium (^{137}Cs) concentrations in water, sediment, aquatic plants and algae, and transfer factors (TF)

* Coexisting

The highest value of water-to-plant TF was 5.37×10^4 (*P. crispus*) and the lowest 6.41×10^2 (*P. distinctus*). The water-to-plant TF of duckweed (a species of *Lemnaceae*) under cultured test conditions was between 2.3×10^3 and 3.9×10^3 (Polar and Bayülgen 1991). The water-to-plant TF of 137 Cs in aquatic plants and algae was between 6.41×10^2 and 5.37×10^4 , which was close to the value reported by Polar and Bayülgen (1991). As filamentous algae and cyanobacteria do not have roots, they obtain the nutrients necessary for growth directly from the water and through vertical movement with gas vesicles, such as *Anabaena* and *Microcystis*, respectively (Ganf and Oliver, 1982). The water-to-plant TF values of filamentous algae and cyanobacteria were on the same order. The soil-to-plant TF values [137 Cs concentration

Bq/kg dry weight_{plant}) × (¹³⁷Cs concentration Bq/kg dry weight_{soil})⁻¹] of terrestrial cyanobacteria *Nostoc commune* contaminated by the FDNPP accident were between 9.8×10^{-1} and 9.59×10 (Sasaki et al. 2013). The water-to-plant TF values of marine macroalgae (seaweed) contaminated by the accident were between ~8 × 10 and 5×10^2 (Kawai et al. 2014). Through screening for useful tools with the potential to decontaminate ¹³⁷Cs, Fukuda et al. (2014) found that algae (a species of *Eustigmatophyceae*) and duckweed (*Lemno aoukikusa*) had high ¹³⁷Cs removal capacities, as determined by culture tests using non-potassium-containing and ¹³⁷Cs-containing media. Potassium and cesium are congeners, i.e., cesium displays similar behavior to that of potassium. Thus, these species of alga and duckweed have the potential to be useful tools

for decontamination. Through this study, we elucidated the behavior of ¹³⁷Cs in aquatic plants and algae in freshwater environments near the FDNPP.

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