

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 ^{137}Cs concentration of $<0.45\text{-}\mu\text{m}$ fractions of water taken from the river and ponds was between 5.01×10^{-1} and 2.98 Bq/L , while that of sediment was between 4.85×10^3 and $5.72 \times 10^4 \text{ Bq/kg}$ dry weight. The ratio of ^{137}Cs concentration of sediment/water in ponds was $\sim 10^4$. The sediment-to-plant transfer factor (TF) [$(^{137}\text{Cs}$ concentration Bq/kg dry weight_{plant}) \times (^{137}Cs concentration Bq/kg dry weight_{sediment}) $^{-1}$] 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 [$(^{137}\text{Cs}$ concentration Bq/kg dry weight_{plant}) \times (^{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 ^{137}Cs concentration of cyanobacteria in pond water was $4.87 \times 10^{-1} \text{ Bq/L}$, which was the same order of magnitude as the ^{137}Cs concentration of pond water. Enrichment of ^{137}Cs in cyanobacteria was not observed.

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 ^{137}Cs 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 ^{137}Cs .

There have been many studies regarding ^{137}Cs transfer in terrestrial plants but few reports on aquatic plants (Avery 1996). Although one report exists regarding the transfer of ^{137}Cs 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, ^{137}Cs concentrations of water, sediment, aquatic plants, and algae in a river and artificial agriculture ponds within $\sim 7 \text{ km}$ 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 ^{137}Cs 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, Niigata, 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 ^{137}Cs 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 water-quality-meter multimonitoring system (W-23XD, HORIBA, 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 ^{137}Cs concentration. The percent moisture content of aquatic plants and algae was determined using the following equation:

$$\text{Percent moisture content} = \frac{(\text{Fresh weight} - \text{Dry weight})}{\text{Fresh weight}} \quad (1)$$

Fig. 1 Sampling points and Fukushima Daiichi Nuclear Power Plant (FDNPP). Aquatic 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 ^{137}Cs concentration

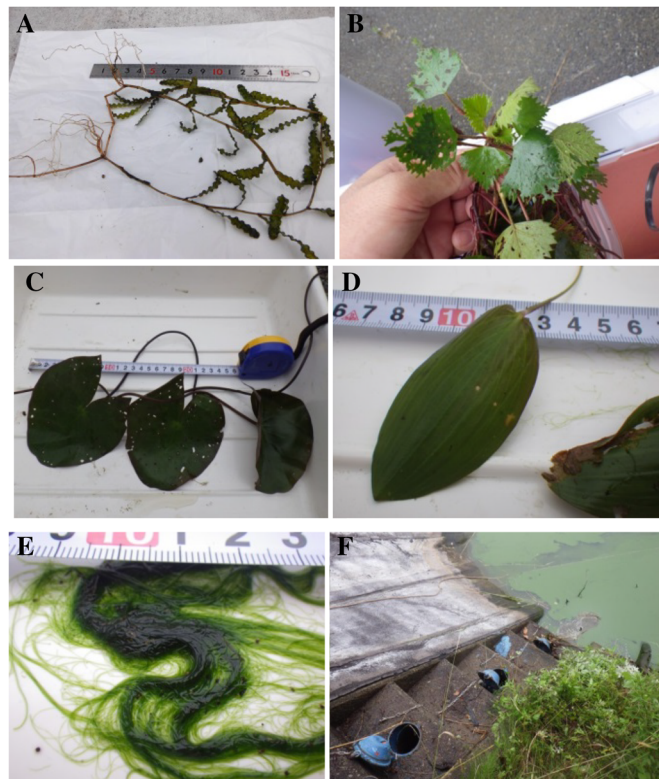
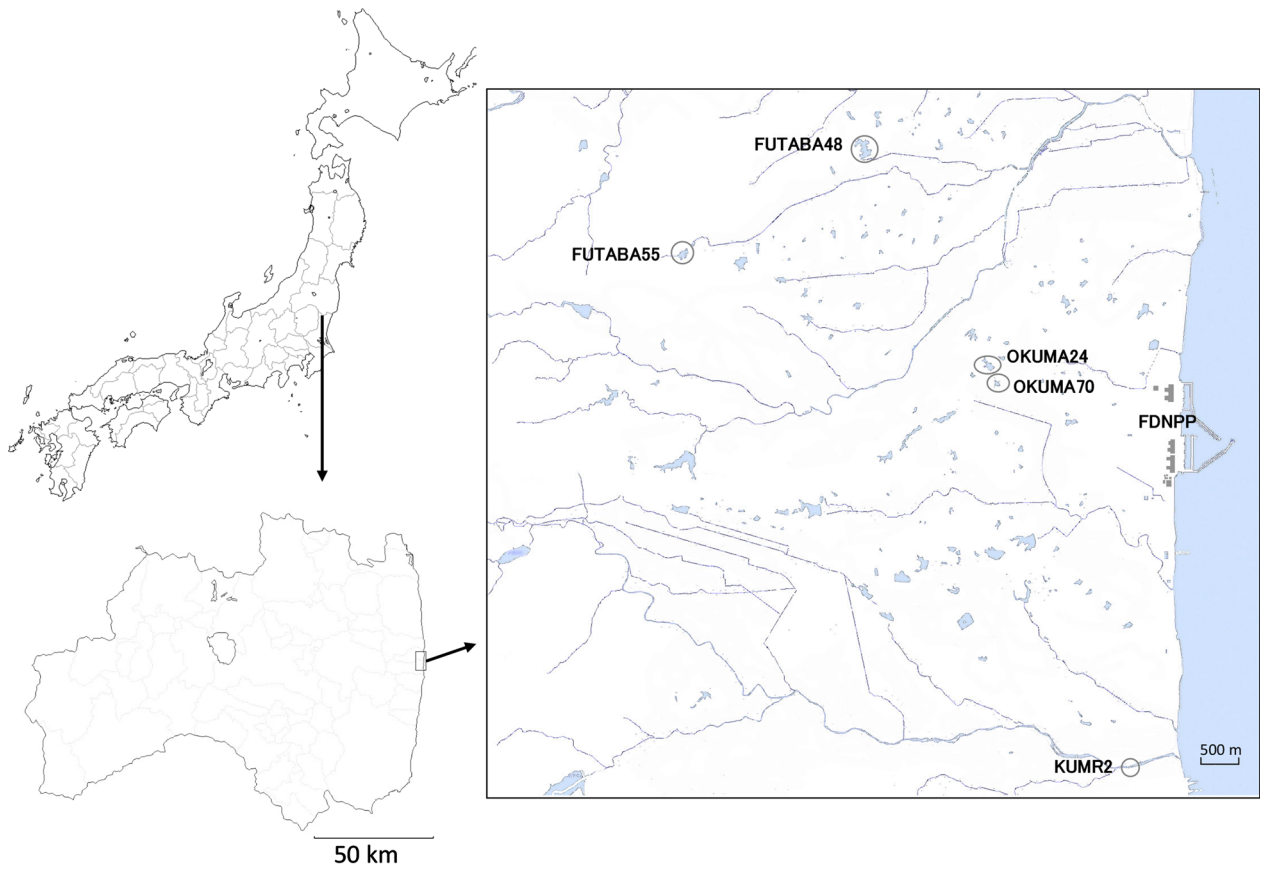
Radiocesium concentrations of water (<0.45-μm fractions in 500-ml plastic containers), dried sediment [in 80-ml plastic (V7) containers], dried aquatic plants (in 100-ml containers), and cyanobacteria (on 47-mm-diameter membrane filters) were measured using an n-type, high-purity Ge-detector (GMX40P4-76 germanium detector, Seiko EG&G ORTEC, Tokyo, Japan) with 40 % relative efficiency. Gamma-ray emission at 604 keV (^{134}Cs) and 661 keV (^{137}Cs) 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 gamma-ray-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, Braunschweig, 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 ^{137}Cs concentration (Bq/kg dry weight), C^w is the water ^{137}Cs concentration (Bq/L), and C^s is the sediment ^{137}Cs concentration (Bq/kg).

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

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



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 ^{137}Cs 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 ^{137}Cs concentration in surface water and sediment is listed in Table 1. The highest concentration of ^{137}Cs 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 ^{137}Cs in river water was estimated by catchment inventory of ^{137}Cs (Santschi et al. 1990, Smith et al. 2004, 2005). The same behavior of ^{137}Cs was observed in the Fukushima River after the FDNPP accident (Yoshimura et al. 2015). The ratio of ^{137}Cs concentration of sediment/water in the ponds was $\sim 10^4$ (Table 1), suggesting that ^{137}Cs concentration in water and sediment is in equilibrium.

Sakaguchi et al. (2015) reported that ^{137}Cs in the river water ($<0.45 \mu\text{m}$ fraction) was present exclusively as the dissolved species rather than being adsorbed on suspended solids or complexed with organic materials. Dissolved ^{137}Cs species in the water are easily incorporated into plants. However, further investigation of the relationship between plant growth and dissolved ^{137}Cs 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 ^{137}Cs concentration of aquatic plants and algae are shown in Table 1. The water content of aquatic plants was $\sim 90\%$, with that of algae being higher. The ^{137}Cs 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 Bq/kg, which was the highest value for the collected aquatic plants. The ^{137}Cs concentration of *Trapa bispinosa* obtained from an adjacent pond, with similar water depth and pH, was similar, as the ^{137}Cs 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 ^{137}Cs 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 ^{137}Cs concentrations of filamentous algae and cyanobacteria were 10^3 Bq/kg. Adsorbed ^{137}Cs on suspended solids of the clay and silt fraction is the main contributor to the transport of ^{137}Cs 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 ^{137}Cs -contaminated microalgae with water flow leads to the potential spread of contamination. The ^{137}Cs 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 ^{137}Cs concentration in the water. In this case, ^{137}Cs migration associated with the movement of cyanobacteria is limited as the ^{137}Cs 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. distinctus* 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 [$(^{137}\text{Cs}$ concentration Bq/kg dry weight_{plant}) \times (^{137}Cs 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 ^{137}Cs , compared with the terrestrial plants.

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

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

* 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 and sediment-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}) \times (^{137}Cs 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 $\sim 8 \times 10$ and 5×10^2 (Kawai et al. 2014). Through screening for useful tools with the potential to decontaminate ^{137}Cs , Fukuda et al. (2014) found that algae (a species of *Eustigmatophyceae*) and duckweed (*Lemma aoukikusa*) had high ^{137}Cs removal capacities, as determined by culture tests using non-potassium-containing and ^{137}Cs -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 ^{137}Cs in aquatic plants and algae in freshwater environments near the FDNPP.

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