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Sediment particle size and initial radiocesium accumulation in ponds following the Fukushima DNPP accident

Kazuya Yoshimura^{1*}, Yuichi Onda¹ & Takehiko Fukushima²Received
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requests for materials
should be addressed toK.Y. (yoshimura.
kazuya@jaea.go.jp);
Y.O. (onda@geoenv.
tsukuba.ac.jp) or T.F.
(fukushima@sakura.cc.
tsukuba.ac.jp)* Current address:
Headquarters of
Fukushima Partnership
Operations, Japan
Atomic Energy
Agency, Fukokuseimei
Building 19F 2-2-2,
Uchisaiwai-cho,
Chiyoda-ku, Tokyo
100-8577, Japan.¹Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan, ²Faculty of Life and Environmental Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan.

This study used particle size analysis to investigate the initial accumulation and trap efficiency of radiocesium (¹³⁷Cs) in four irrigation ponds, ~4–5 months after the Fukushima Dai-ichi nuclear power plant (DNPP) accident. Trap efficiency, represented by the inventory of ¹³⁷Cs in pond sediment to the inventory of radiocesium in soil surrounding the pond (i.e., total ¹³⁷Cs inventory), was less than 100% for all but one pond. Trap efficiency decreased as sediment particle size increased, indicating that sediments with a smaller particle size accumulate more ¹³⁷Cs. In ponds showing low trap efficiency, fine sediment containing high concentrations of ¹³⁷Cs appeared to be removed from the system by hydraulic flushing, leaving behind mostly coarse sediment. The results of this study suggest that sediment particle size can be used to estimate the initial accumulation and trap efficiency of ¹³⁷Cs in pond sediment, as well as the amount lost through hydraulic flushing.

Contamination of reservoir sediment with radiocesium has been thoroughly investigated, particularly since the accident at the Chernobyl nuclear power plant. Temporal changes in ¹³⁷Cs concentration, inventory^{1–4}, spatial/vertical distribution^{5–9}, and physicochemical properties^{10–12} have been well documented in reservoirs. However, few studies have documented the initial accumulation and flushing of ¹³⁷Cs from reservoirs exposed to the fallout of radioactive materials from nuclear power plant accidents.

Reservoir inputs of ¹³⁷Cs are removed from the water column mainly by direct adsorption to sediment^{1,13,14}, sedimentation with suspended particles^{12,15}, and hydraulic flushing¹. Davison *et al.*¹ investigated the concentration of ¹³⁷Cs in the water and sediments of English lakes immediately following the Chernobyl nuclear power plant accident. They estimated, from the flow rate and concentration of radiocesium in the water, that 32–41% of the ¹³⁷Cs that was initially deposited had been hydraulically flushed from the lakes, and the remaining ¹³⁷Cs had accumulated in sediments. However, it is difficult to collect such data from many reservoirs immediately following a nuclear accident.

The concentration of ¹³⁷Cs in reservoir sediment can be affected by many factors, such as the initial amount of radiocesium deposition, organic matter content of the sediment, water depth, catchment area, pond volume, and hydraulic residence time^{2,3}. Additionally, it has been well established that sediment particle size can affect the concentration of ¹³⁷Cs in sediment^{10,16–19}. He and Walling¹⁰ demonstrated, by laboratory experiments and empirical observation, that the increase in ¹³⁷Cs concentration in soil and sediment was associated with a decrease in particle size. Radiocesium in the surface water of ponds, which are shallower than lakes and usually only a few meters deep, is inferred to be directly associated with resuspended solids, particularly fine particles. Since hydraulic flushing of fine particles is reflected in the particle size distribution of the sediment, assessment of sediment particle size is important in determining the amount of ¹³⁷Cs removed from a reservoir by hydraulic flushing.

On March 12, 2011, an accident at the Fukushima Dai-ichi nuclear power plant (DNPP) released ~1–2 × 10¹⁶ Bq of ¹³⁷Cs into the atmosphere²⁰, contaminating a large proportion of the surrounding area^{21–24}, which included a forested area²⁵. Fukushima Prefecture, the area most affected by the ¹³⁷Cs fallout, is a major rice-farming region where there are many artificial ponds used for irrigation, which are surrounded by forest. Since reservoirs accumulate soil eroded from the catchment and decrease the flux of sediment to downstream systems²⁶, it is also likely that they have an impact on the movement of sediment-sorbed radiocesium to downstream systems. It has been reported that the initial inventory of ¹³⁷Cs in lakes is related to the total ¹³⁷Cs directly deposited in/around the lakes, with a small contribution of ¹³⁷Cs from the catchment^{1,13}. Thus, trap efficiency can be calculated as the proportion of ¹³⁷Cs inventory in pond sediment to the ¹³⁷Cs inventory in the soil

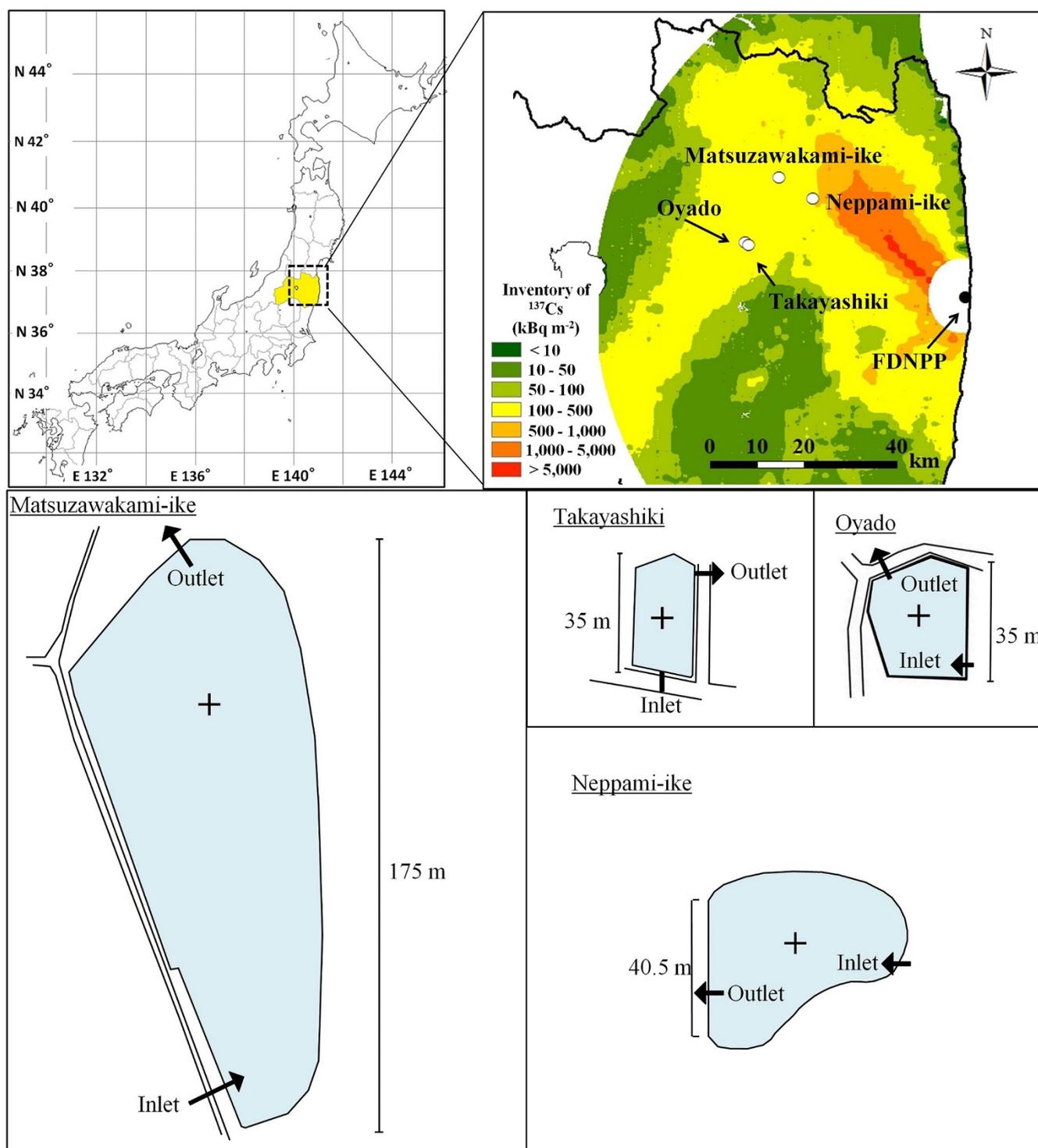


Figure 1 | Location and plain views of irrigation ponds. Cross in plain view indicates sampling point. These maps were generated by ArcGIS 10 software and combined with the ^{137}Cs distribution obtained from the Third Airborne Monitoring Survey (MEXT²¹), provided in the Database on the Research of Radioactive Substances Distribution³⁴.

surrounding the pond (i.e., total ^{137}Cs inventory). Assessing the accumulation and flushing of ^{137}Cs in ponds is important in understanding the initial contamination of sediment and the discharge of radiocesium to downstream systems. To our knowledge, no studies have investigated the initial dynamics of ^{137}Cs in irrigation pond sediment following the Fukushima DNPP accident.

In this study, we assessed the concentrations of ^{137}Cs in the sediments of four irrigation ponds (Fig. 1) 4–5 months after the

Fukushima DNPP accident. We used sediment particle size analysis as a simplified estimation of ^{137}Cs dynamics, to determine initial accumulation and hydraulic flushing of ^{137}Cs in ponds exposed to nuclear fallout.

Results

The description of each pond, sampling dates, and ^{137}Cs inventory in the soil surrounding the ponds, which was measured as part of the



Table 1 | Descriptions of ponds, sampling dates, and inventories of ^{137}Cs in soils surrounding ponds estimated from the Third Airborne Monitoring Survey, by MEXT²¹. Total inventories of ^{137}Cs were estimated as inventory on ground surrounding the ponds within 25 m distance, and the decay is corrected on 2 Jul. 2011

Descriptions	Oyado	Takayashiki	Neppami-ike	Matsuzawakami-ike
Surface area (m ²)	804	673	2400	8720
Maximum depth (m)	0.7	0.3	1.4	2.0
Catchment area (ha)	3.6	1.6	20	9.8
Sampling date	8 Jul. 2011	8 Jul. 2011	4 Aug. 2011	4 Aug. 2011
Total inventory of ^{137}Cs (kBq m ⁻²)	360	318	349	206

Third Airborne Monitoring Survey by MEXT on 2 July 2011 (calibrated with 2200 points of soil core samples²¹), are provided in Table 1. The inventory of ^{137}Cs in the surrounding soil was estimated to be 206–360 kBq m⁻². While heterogeneity in the local distribution of radioactivity has been observed²⁷, no hot particles derived from the fuel of nuclear reactor have been detected around our sites²⁸.

The mass-depth distributions and inventories of ^{134}Cs and ^{137}Cs in the pond sediments are presented in Fig. 2. In all ponds, the radio-cesium concentrations were highest in the surface layer of sediment and quickly decreased with depth. Concentrations of ^{137}Cs in the surface layer were 17.9, 34.9, 6.4, and 10.9 kBq kg⁻¹ at Oyado, Takayashiki, Netsupami-ike, and Matsuzawakami-ike, respectively. Radio-cesium was detected as far down as the fifth layer at Takayashiki, and the fourth layer in the other ponds. However, >80% of the total ^{134}Cs and ^{137}Cs inventories were in the top two surface layers (<16 kg m⁻²). The accumulation of ^{137}Cs in surface sediment (<3.5 cm) was simulated in laboratory experiments, using the kinetics of ^{137}Cs sorption to and desorption from sediments^{29,30} and the vertical distribution of ^{137}Cs in our sediment samples. The results indicate that downward migration or agitation of radio-cesium in sediment was minimal in the ponds studied, and that most of the radio-cesium was trapped in surface sediment.

Matsuzawakami-ike has a relatively large surface area compared to the other ponds (Fig. 1; Table 1), so there could be spatial variation in the inventory of radio-cesium in sediment. Another investigation collected sediment core samples at three locations (inlet-side, middle, and outlet-side) in Matsuzawakami-ike at 30–40 m intervals in Apr. 2013 (not published). The coefficient of variation for ^{137}Cs -inventory was 7%; thus, the spatial variation in inventory was inferred to be small.

Radio-cesium inventory was related to the concentrations of radio-cesium in sediment; the highest radio-cesium inventory was observed in Takayashiki, which showed the highest concentration of radio-cesium. Radio-cesium inventory in Oyado, which had the highest amount of ^{137}Cs deposition among the study ponds, was lower than that in Takayashiki, indicating a difference in radio-cesium accumulation efficiency. To evaluate the efficiency of sediment in accumulating radio-cesium, we compared the inventory of ^{137}Cs in pond sediment to the total ^{137}Cs inventory (Table 1), referred to hereafter as trap efficiency. Based on the findings of previous studies^{1,13}, we assumed that the amount of ^{137}Cs deposited in the surrounding soils represented the total amount of ^{137}Cs deposited into the ponds. There was marked variation in trap efficiency among ponds: 23, 133, 19, and 37% for Oyado, Takayashi, Netsupami-ike, and Matsuzawakami-ike, respectively.

Reservoir characteristics, such as water depth, catchment area, and pond volume, can affect the hydraulic residence time and the associated removal of ^{137}Cs through hydraulic flushing. The ratio between catchment area and pond volume was calculated as an estimate of hydraulic residence time, which was compared to the trap efficiency. There was no significant correlation between our estimate of hydraulic residence time and the trap efficiency. We also investigated the elemental composition, geology, and organic content of sediment, which are factors that can affect the adsorption of radio-cesium

by sediment particles. The elemental composition is presented in Table 2. There was no unusual composition, and there was no clear relationship between content and trap efficiency for any sediment element. As regards the geology of the ponds studied, they are all granite bedrock³¹. The average organic matter contents in the three surface layers of sediment were $16.7 \pm 0.6\%$, $12.6 \pm 0.1\%$, $12.0 \pm 0.2\%$, and $12.3 \pm 1.0\%$ for Takayashiki, Oyado, Netsupami-ike, and Matsuzawa-ike, respectively, and there was no significant correlation between organic matter content and trap efficiency. Water quality factors such as pH can also affect the adsorption/desorption dynamics of radio-cesium in sediment. The pH in each pond was measured in April and May 2013. The pH values were 7.5–7.7 for Takayashiki, 7.5–7.6 for Oyado, 7.6–7.9 for Netsupami-ike, and 7.9–8.0 for Matsuzawakami-ike (personal data); there was no correlation between pH and trap efficiency. Based on these results, the variation in trap efficiency was not the result of differences in the reservoir characteristics, the sediment characteristics, or pH among study ponds.

The inventories of radio-cesium reflected the concentration of radio-cesium in our study ponds. It has been established that sediment particle size can affect the concentration of radio-cesium in sediment^{10,16–19}. Therefore, we assessed the effect of particle size by comparing ^{137}Cs -based weighted average values of D_{50} and specific surface area to trap efficiency (Fig. 3). The D_{50} and specific surface area of sediment in Takayashiki, which had the highest trap efficiency among our study ponds, were 15.1 μm and 0.41 m² g⁻¹, respectively, representing the smallest particle size among ponds. Trap efficiency was correlated with sediment particle size, showing a negative correlation with D_{50} ($r^2 = 0.69$; Fig. 3a) and a positive correlation with specific surface area ($r^2 = 0.66$; Fig. 3b).

Discussion

Davison *et al.*¹ reported trap efficiencies between 59–68% for two English lakes, over an 18-month period after the Chernobyl nuclear power plant accident, and that 32–41% of the total ^{137}Cs was hydraulically flushed from the system. It is likely then, for the ponds in this study that had trap efficiencies <100%, that some of the initial radio-cesium inputs were removed through hydraulic flushing. Hydraulic flushing of ^{137}Cs from a reservoir can be affected by precipitation, as large rain events will lead to shorter hydraulic residence times. Oyado and Takayashiki are very close to each other (<1 km apart) and likely experience similar rates of precipitation, but they had very different trap efficiencies (23 vs. 133%). While reservoir characteristics, such as water depth, catchment area, pond volume, and hydraulic residence time can also affect hydraulic flushing^{2,3} of ^{137}Cs from ponds, we observed no correlation between these factors and trap efficiency. However, trap efficiency was correlated with sediment particle size, indicating that particle size can be an important factor in determining trap efficiency. Accumulation of ^{137}Cs in sediment occurs through direct adsorption to sediment^{1,13,14} and sedimentation with suspended particles^{12,15}. It is likely that the finer sediment in Takayashiki facilitated the accumulation of ^{137}Cs , resulting in the highest trap efficiency. In contrast, ponds with coarser sediment and lower trap efficiencies may have lost ^{137}Cs through

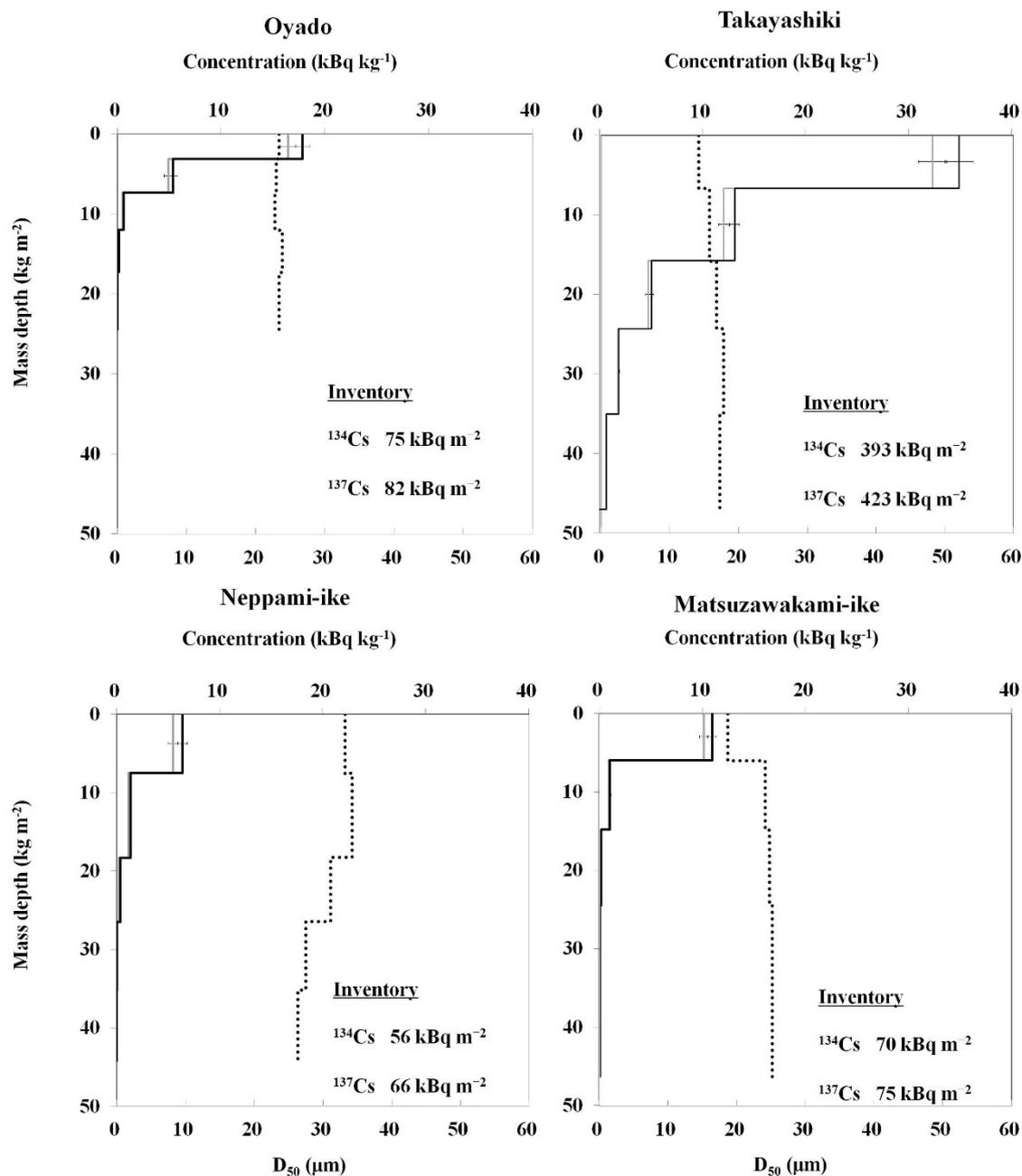


Figure 2 | Mass depth distributions of concentrations of radiocesium in sediment (gray line for ¹³⁴Cs and black line for ¹³⁷Cs) and D₅₀ of sediment (broken line). Inventories are also shown.

hydraulic flushing of finer sediment. Thus, sediment particle size is one of important factors in estimating the amount of ¹³⁷Cs lost through hydraulic flushing, in addition to its accumulation in pond sediment. More research and data are needed to determine the strength of the correlation between radiocesium trap efficiency and sediment particle size.

Methods

Sample collection. We selected four irrigation ponds, located ~40–50 km from the Fukushima DNPP (Fig. 1), to investigate initial ¹³⁷Cs accumulation from the fallout of the Fukushima DNPP accident. Oyado and Takayashiki are located in the city of Nihonmatsu, and Netsupami-ike and Matsuzawakami-ike are located in the town of Kawamata. The ponds were artificially constructed (i.e., reservoirs) and receive water inputs from precipitation and streams within the

Table 2 | Elemental composition of surface (0–2 cm layer) sediments (%). The value was obtained as weight composition

Site	Fe	Mn	Ti	Ca	K	Al	Si
Takayashiki	29	1	3	6	9	10	42
Oyado	34	0	4	6	7	8	41
Netsupami-ike	35	1	2	6	11	6	39
Matsuzawakami-ike	36	1	4	8	8	8	36

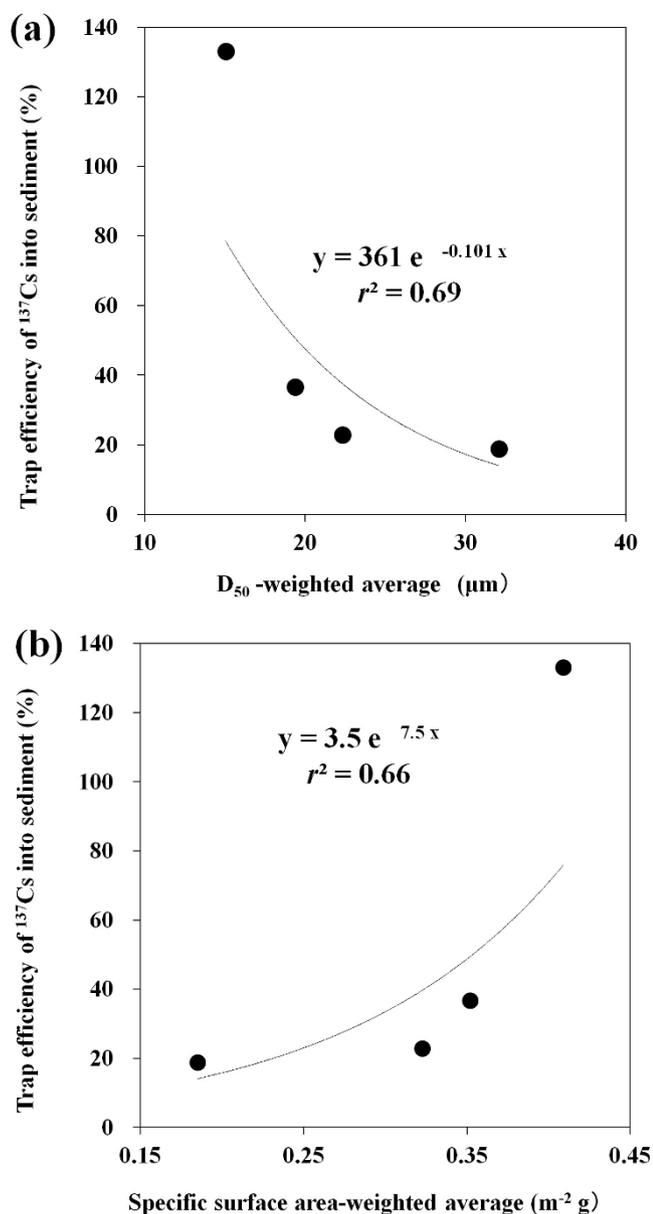


Figure 3 | Variation in trap efficiency of ^{137}Cs into sediments associating with weighted averages of (a) D_{50} and (b) specific surface area. Solid curves indicate regression lines.

catchment. As the ponds all have relatively flat bottoms, water depths were similar for large areas in all of the ponds.

Sediments were collected at Oyado and Takayashiki on 8 July 2011, and at Netsupami-ike and Matsuzawakami-ike on 4 August 2011. Four sediment cores (core depth: >12 cm) were taken from approximately the center of each pond using a gravity corer (i.d.: 4 cm). Each core was sampled at a distance of 1–3 m from other core-sampling points to avoid the influence of sediment agitation on samples. Core samples were sliced into 2-cm layers, for a total of five layers for each core. Across replicate cores, sliced samples were combined at each depth. The samples were then dried at 105°C for 24 h. The dried samples were homogenized, using a mortar and pestle, and used for measurements.

Sample analysis. Concentrations of ^{137}Cs and ^{134}Cs were measured using a high-purity n-type germanium coaxial gamma-ray detector (EGC25–195–R, Canberra–Eurisys, Meriden, U.S.A.), equipped with an amplifier (PSC822, Canberra, Meriden, U.S.A.) and multichannel analyzer (DSA1000, Canberra, Meriden, U.S.A.). Instruments were calibrated using standard gamma sources: ^{210}Pb and ^{137}Cs (EG-CUSTOM; Isotope Products Laboratories, USA), ^{241}Am , ^{109}Cd , and ^{152}Eu (EG-CUSTOM; Eckert and Ziegler Isotope Products, USA). The samples were placed on top of the detector head and radioactivity was measured for 5 min to 12 h, depending on the activity of the sample. The activities of ^{137}Cs and ^{134}Cs were corrected for decay, based on when the samples were collected. The counting error was less than 10%, and

the measurement accuracy satisfied the IAEA–CU–2006–03 World–Wide Proficiency Test on the Determination of Gamma Emitting Radionuclides³². Further details can be found in Kato *et al.*³³.

Samples were then analyzed using a laser diffraction particle size analyzer (SALD–3100, Shimadzu Co., Ltd., Kyoto, Japan) to determine the median-diameters (D_{50}) and specific surface areas of sediment particles, for each layer of sediment. To evaluate the effect of particle size on the accumulation of ^{137}Cs in sediment, ^{137}Cs -based weighted average values of D_{50} and specific surface area were calculated according to the following equation:

$$\text{Weighted average } V = \frac{\sum_{i=1}^5 I_i V_i}{I_{\text{total}}} \quad (1)$$

where V_i is D_{50} or specific surface area, I_i is the inventory of ^{137}Cs in layer i , and I_{total} is the total inventory of ^{137}Cs in a given sediment core.

After radiocesium analysis, the elemental composition of each surface layer of sediment (0–2 cm) was determined using an X-ray fluorescence analyzer (Niton XL3t, Thermo Scientific) with a helium gas-purging system. To measure the organic content, ~1 g of sample was subdivided from three surface layers (0–2-, 2–4-, and 4–6-cm layers). The subdivided samples were weighed, and then combusted at 500°C for 24 h. The mass lost upon ignition represented the organic content.

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Author contributions

K.Y. and T.F. carried out the sampling. K.Y. performed measurement and analysis. K.Y. and Y.O. discussed the results and contributed to the manuscript preparation.

Additional information

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