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Contents lists available at ScienceDirect

Chemosphere



journal homepage: www.elsevier.com/locate/chemosphere

Radiocesium distribution and mid-term dynamics in the ponds of the Fukushima Dai-ichi nuclear power plant exclusion zone in 2015–2019

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HIGHLIGHTS

• ¹³⁷Cs was persistent in ponds of the Fukushima exclusion zone in 2015 –2019.

- Conceptual model for transformation of ¹³⁷Cs speciation was adapted for Fukushima.
- Apparent $K_d(^{137}Cs)$ declined with time in ponds with rate constant 0.12 -0.18 yr^{-1} .
- Exchangeable radiocesium interception potential of sediments was, on the average, 2050 mEq/kg.

ARTICLE INFO

Article history: Received 31 August 2020 Received in revised form 16 November 2020 Accepted 18 November 2020 Available online xxx

Handling Editor: Martine Leermakers

Keywords: Fukushima Pond Radiocesium Dissolved Particulate Leaching

GRAPHICAL ABSTRACT



ABSTRACT

This study analyzes the ¹³⁷Cs behavior in the ponds of Okuma Town from 2015 to 2019 in the Fukushima Dai-ichi nuclear power plant (FDNPP) exclusion zone. A decline in both particulate and dissolved ¹³⁷Cs activity concentrations was revealed. The decline rate constants for the particulate ¹³⁷Cs activity concentration were found to be higher than for the dissolved ¹³⁷Cs activity concentration. In terms of seasonality the dissolved ¹³⁷Cs concentrations were higher from June to October, depending on the specific pond and year, most likely due to temperature dependence of ¹³⁷Cs desorption from frayed edge sites of micaceous clay minerals. The apparent $K_d(^{137}Cs)$ in the studied ponds, in absolute value, appeared to be much higher than that for closed and semi-closed lakes of the Chernobyl contaminated area; however, these were comparable to the values characteristic of the rivers and reservoirs of the FDNPP contaminated area. The apparent $K_d(^{137}Cs)$ in the suspended sediment–water system was observed to decrease over time. It was hypothesized that this trend was associated with the decomposition of glassy hot particles. Relying on the theory of selective sorption and fixation, the exchangeable radiocesium interception potential, $RIP^{ex}(K)$ was estimated using data on ¹³⁷Cs speciation in the surface bottom-sediment layer and its distribution in the sediment–water system. For the studied ponds, $RIP^{ex}(K)$ was on the

Abbreviations: FDNPP, Fukushima Daiichi nuclear power plant; NPP, nuclear power plant; MEXT, Ministry of Education, Culture, Sports, Science and Technology; RES, regular surface ion-exchange sites; FES, frayed edge ion-exchange sites; CsMP, Cs-rich microparticles.

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https://doi.org/10.1016/j.chemosphere.2020.129058

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Please cite this article as: A. Konoplev, Y. Wakiyama, T. Wada *et al.*, Radiocesium distribution and mid-term dynamics in the ponds of the Fukushima Dai-ichi nuclear power plant exclusion zone in 2015–2019, Chemosphere, https://doi.org/10.1016/j.chemosphere.2020.129058

average 2050 mEq/kg, which is within the range of values measured in laboratory studies reported in the literature.

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1. Introduction

The Fukushima Dai-ichi nuclear power plant (FDNPP) accident caused by the tsunami following the Great East Japan Earthquake in March 2011 resulted in the contamination of extensive areas of Honshu Island (Japan) by radioactive cesium isotopes ^{134}Cs (half-life $T_{1/2}\,=\,2.06$ years) and ^{137}Cs ($T_{1/2}\,=\,30.17$ years). This has rekindled the interest in the behavior of radiocesium, particularly given the geoclimatic conditions of Japan. Radiocesium was deposited north-west of the nuclear power plant (NPP), forming a footprint approximately 20 km wide and 50-70 km long (Chino et al., 2011; Hirose, 2012; Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2012) and leading to the contamination of both terrestrial and aquatic ecosystems. The initial activity ratio of cesium isotopes ¹³⁴Cs/¹³⁷Cs in the Fukushima fallout was close to unity (Hirose, 2012). With time, the contribution of ¹³⁴Cs to radioactive contamination of the environment decreased relative to ¹³⁷Cs due to its faster decay, and ¹³⁷Cs is currently the main radionuclide of dose significance.

Right after the FDNPP accident, the hypothesis was that radiocesium had deposited as part of condensation particles in watersoluble forms (Kaneyasu et al., 2012) assuming that radiocesium was transported in the atmosphere by sulfate aerosol particles of 0.5-0.6 µm diameter, radiocesium in these particles to be watersoluble and washable by precipitation. However, later studies (Adachi et al., 2013; Abe et al., 2015) have revealed spherical glassy aerosol particles of a few µm in diameter, as far as 170 km from the FDNPP, containing, apart from radiocesium, uranium and other elements representative of reactor materials. Particles of similar properties have also been identified by Niimura et al. (2015) using autoradiography of soils, plants and mushrooms. Later on, coarser particles (up to hundreds of µm) that have higher radiocesium activity (sometimes more than kBq per particle) with irregular shape have been identified in surface soil and dust samples collected in the vicinity of FDNPP within a few km range (Igarashi et al., 2019). The major component of these radiocesium-bearing microparticles (CsMPs) is SiO₂ (Satou et al., 2018). What is important, CsMPs are insoluble in water and persistent in the environment (Igarashi et al., 2019; Miura et al., 2020). In terms of the behavior of Fukushima-derived radiocesium the critical questions regarding CsMPs are: 1) what was the fraction of CsMPs activity in the total radiocesium release and deposition at different locations? and 2) what is the rate of radiocesium leaching from CsMPs as a result of their decomposition?

Ikehara et al. (2018 and 2020) succeeded to characterize quantitatively the content of CsMPs in surface soils collected in 20 locations of the contaminated area at different directions from the FDNPP. They found that CsMPs account for a significant fraction of Fukushima-derived radiocesium. Based on the plume trajectories they concluded that CsMPs were formed during a relatively short period from the late afternoon of March 14, 2011 to the late afternoon of March 15, 2011, and that Unit 3 of the FDNPP was the most plausible source of the CsMPs at the beginning of the release. The number of CsMPs in surface soils at various directions, distances and deposition levels in the soils was in the range 0.9–101 particles/g, and the radioactivity fraction of CsMPs ranged in 15–80%. Occurrence of CsMPs in soils and sediments can substantially impact the radiocesium solid-liquid distribution in the soil-water environment (Konoplev, 2015). However, the variation of CsMPs fraction in deposition in a wide range makes it difficult to account for CsMPs in assessment of mobility and bioavailability of radiocesium in the soil-water environment and its dynamics (Reinoso-Maset et al., 2020).

Miura et al. (2018) discovered CsMPs in suspended sediments collected from Kuchibuto River (right bank tributary of the Abukuma River flowing from the most contaminated section of the Abukuma River catchment) during 2011–2016. The fraction of radiocesium incorporated in CsMPs to the total radiocesium in the sediments was found to be up to 67%. Okumura et al. (2019) showed in laboratory experiments with individual CsMPs that they can be dissolved by weathering in the environment with a rate dependent on temperature and water composition. Temperature dependence of CsMP dissolution was characterized by activation energy 65 kJ/ mol for pure (deionized) water and 88 kJ/mol for seawater (Okumura et al., 2019).

After deposition on the surface of ground and water bodies radiocesium becomes incorporated in physicochemical and biological processes, which lead to changes in its speciation (Beresford et al., 2016; Konoplev, 2020; Salbu et al., 2004, 2018). Glassy hot particles get disintegrated, and radiocesium incorporated in these particles transfers to solution (Konoplev et al., 2018). Dissolved radiocesium is adsorbed on the solid phase by ion exchange (Cremers et al., 1988; Konoplev and Bulgakov, 2000). Exchangeable radiocesium transforms into nonexchangeable (fixed) form as a result of replacement of interlattice K-cations by Cs-cations due to collapse of expanded edges of clay mineral's crystal lattice interlavers (Beresford et al., 2016: Konopley, 2020; Sawhney, 1972). Data for long-term transformation of chemical forms of radiocesium in soil after nuclear weapon testing (Pavlotskaya, 1974), the Kyshtym accident (Konoplev and Bobovnikova, 1991) and 34 years after the Chernobyl accident indicate the existence of a remobilization process that is the reverse of fixation (Konoplev et al., 1992a; Smith and Comans, 1996; Konoplev and Bulgakov, 2000). The fraction of radiocesium exchangeable form does not decrease to zero, as is supposed to happen due to irreversible fixation, but it decreases to a certain stationary level, and then does not change significantly because of establishing a steady state between fixation and remobilization (Beresford et al., 2016; Konoplev and Bobovnikova, 1991; Konoplev, 2020). The key processes of transformation of Fukushima-derived radiocesium chemical forms in the sediment-water environment are shown in Fig. 1.

The partitioning of the radionuclide between the sediment and solution is usually described by the apparent distribution coefficient K_d (L/kg) defined as the ratio of the particulate radionuclide activity concentration $[R]_p$ (Bq/kg) to its dissolved activity concentration $[R]_d$ (Bq/L) at equilibrium (IAEA, 2010):

$$K_d = \frac{[R]_p}{[R]_d} \tag{1}$$

For contaminated areas of Fukushima, $[R]_p$ includes the radiocesium embedded in glassy hot microparticles, the exchangeably sorbed radiocesium, and radiocesium fixed by clay minerals in sediments (Fig. 1). The exchangeable form of radiocesium occurs at



Fig. 1. Conceptual model of transformation processes for Fukushima-derived radiocesium speciation in sediment-water system (modified after Konoplev et al., 2018).

instantaneous ion-exchange equilibrium with the liquid phase, whereas the non-exchangeable form is not involved in the radiocesium exchange with the solution in the immediate term. The concept of the exchangeable distribution coefficient K_d^{ex} , which is the ratio of the exchangeable radionuclide activity concentration in sediments $[R]_{ex}$ to its activity concentration in solution at equilibrium $[R]_d$, is widely used now (Konoplev et al., 1992a; Konoplev and Bulgakov, 2000; Smith et al., 2000):

$$K_d^{ex} = \frac{[R]_{ex}}{[R]_d} = \alpha_{ex} K_d \tag{2}$$

where α_{ex} is the fraction of the exchangeable radionuclide in sediments, and K_d is the apparent (also known as total) distribution coefficient.

It is now commonly believed that the increased ability of soils and sediments to selectively sorb radiocesium is due to the presence of micaceous clay minerals (Beresford et al., 2016; Cremers et al., 1988; Konoplev, 2020; Sawney, 1972; Vandebroek et al., 2012). At least two types of sorption sites can be distinguished: regular surface ion-exchange sites (RES) and highly selective sorption sites for cesium occurring in the area of the fraved edges of neighboring layers of micaceous clay crystal lattice (FES). Radiocesium is adsorbed on RES non-selectively, i.e., the selectivity coefficient of its sorption with respect to one-charge ions K⁺, Na⁺, NH⁺₄ and others is close to one. In contrast, the selectivity coefficient for Cs sorption on FES is approximately 1000 for K⁺ and approximately 200 for NH₄⁺ (Wauters et al., 1996a). The FES (selective sorption sites) constitute a relatively small portion of ionexchange sorption sites (1%–5%) for most soils and sediments (De Preter, 1990). Due to high selectivity of FES for cesium and because radiocesium (and even stable cesium) occurs at trace concentrations in the environment, the exchangeable radiocesium is almost entirely adsorbed by FES in most sediments and soils.

Using the exchangeable distribution coefficient has certain merits because this parameter is governed by physically meaningful characteristics of the environment such as the capacity of sorption sites in sediments and water cation composition (Konoplev, 2020; Konoplev and Bulgakov, 2000; Konoplev et al., 2002; Ries et al., 2019; Smith et al., 2000). Conversely, the value of the apparent K_d is strongly dependent on the non-exchangeable radiocesium fraction which is determined by its initial speciation in the fallout and the rates of radiocesium leaching from glassy hot particles (k_l) , fixation (k_f) , and remobilization (transformation of fixed form to exchangeable form) (k_r) (Fig. 1). The laboratory experiments with various soils and sediments (Konoplev et al., 1996) and post-Chernobyl field studies (Konoplev et al., 1992a) have shown that the timescale of radiocesium fixation is weeks or months, depending on environmental conditions, while the timescale of remobilization can be up to a few years. However, the rate of radiocesium leaching from CsMP has not been reliably determined for environmental conditions yet. It can be expected that this process in the contaminated areas of Fukushima is relatively slow, even slower than decomposition and radionuclide leaching from Chernobyl-derived fuel particles (Beresford et al., 2016; Bobovnikova et al., 1991; Kashparov et al., 1999, 2020; Konoplev and Bobovnikova, 1991; Konoplev, 2020; Konoplev et al., 1992a) since glassy particles may prove to be more persistent in the environment than UO₂ fuel particles when exposed to oxidation by atmospheric oxygen.

The ability of sediment to sorb radiocesium selectively is described in terms of the capacity of the selective sorption sites (FES) or the so-called radiocesium interception potential (RIP), which is the product of the FES capacity and the selectivity coefficient of radiocesium in relation to the corresponding competitive ion (Cremers et al., 1988). Soon after the Chernobyl accident a method for quantitative determination of FES capacity ([FES]) and RIP was developed (Cremers et al., 1988; De Preter, 1990; Sweeck et al., 1990). It was then essentially simplified (Wauters et al., 1996b), and later modified within the concept of exchangeable distribution coefficient K_d^{ex} (Konoplev and Konopleva, 1999; Konoplev et al., 2002). From this perspective, the exchangeable radiocesium interception potential *RIP*^{ex}(*K*) is defined as follows:

$$RIP^{ex}(K) = K_c(Cs/K)[FES] = K_d^{ex}[K^+]$$
(3)

where $K_c(Cs/K)$ is the selectivity coefficient for ion exchange of Cs^+ on FES with respect to cation K^+ . $RIP^{ex}(K)$ is the intrinsic property of a given sediment that characterizes its ability to sorb cesium selectively and reversibly. In mixed cases, when the relative contributions of potassium and ammonium to competition with Cs are comparable, the following expression is applicable (Konoplev, 2020; Konoplev et al., 2002; Wauters et al., 1996a, 1996b):

$$RIP^{ex}(K) = K_d^{ex}\left([K^+] + K_c^{FES}(NH_4 / K)[NH_4^+]\right)$$

$$\tag{4}$$

where $K_c^{FES}(NH_4/K)$ is the selectivity coefficient of ammonium with respect to potassium for the FES sites. As shown by the post-Chernobyl studies, most soils and sediments are characterized by $K_c^{FES}(NH_4/K) \approx 5$, hence equation (4) can be rewritten as follows (Konoplev et al., 2002; Wauters et al., 1996a):

$$K_d^{ex} = \frac{RIP^{ex}(K)}{[K^+] + 5[NH_4^+]}$$
(5)

Typical soils of Fukushima contaminated areas are characterized by the relatively high content of clay minerals (up to 30% or more) and a sufficient amount of micaceous clay minerals (Akai et al., 2013; Nakao et al., 2014, 2015a; Yamaguchi et al., 2017, 2019). In the soils from the eastern (Hama-dori) and western (Aizu) parts of Fukushima Prefecture smectite was found to be the predominant clay mineral, while being variable for soils from the central part (Naka-dori) (Nakao et al., 2014). The content of potassium in soil clavs is thought to be an indicator of the amount of micaceous minerals. (Nakao et al., 2014, 2015a). The RIP theory and methodology developed after the Chernobyl accident was widely used to characterize soils and sediments of Fukushima contaminated areas in terms of their ability to adsorb and fix radiocesium (Fan et al., 2014; Fujii et al., 2019; Kogure et al., 2012; Mukai et al., 2014; Mukai et al., 2016a; Mukai et al., 2016b; Nakao et al., 2008; Nakao et al., 2015b; Nakao et al., 2019; Okumura et al., 2018). Unfortunately, radiocesium fixation by clay minerals was not taken into account by the protocol used in most of these works, and therefore obtained data could overestimate RIP^{ex} and underestimate RIP at saturated fixation (Konoplev and Konopleva, 1999; Konoplev et al., 2002). Nakao et al. (2015a) determined RIP for 97 paddy soils from Fukushima Prefecture representing a variety of soil types in the region. The mean value of RIP for 97 studied soils was 1670 \pm 870 mEq/kg and ranged from 340 to 5360 mEq/kg. Yamaguchi et al. (2017) measured RIP for 925 farmland soil samples collected from Fukushima Prefecture and surrounding regions. RIP values ranged from 73 to 12,700 mEq/kg, with wide range observed among samples with identical soil types and geological features. Since topsoil is the main source of particulate matter for water bodies in the surrounding areas (Konoplev et al., 2021), sediments can be expected to be characterized by similar properties. The RIP values obtained for Fukushima soils are compatible with those for 120 European soils and sediments obtained by the same protocol (Wauters et al., 1996b).

Studies on various radioactively contaminated areas over the world have shown that closed and semi-closed water bodies such as lakes and ponds are more sensitive to contamination with radiocesium, i.e. ¹³⁷Cs is much more persistent in abiotic and biotic compartments in these water bodies than in rivers, dam reservoirs and large open lakes (Mokrov et al., 2008; Pinder et al., 2010; Strand et al., 1999; Whicker et al., 1990). In numerous post-Chernobyl studies, elevated mobility and bioavailability of radiocesium was detected in closed and semi-closed lakes and ponds as compared with rivers (Bulgakov et al., 2002; Comans et al., 1989, 1998; Kanivets et al., 2020; Konoplev et al., 1992b, 1998, 2002; Ries et al.,

2019; Zibold et al., 2002). Therefore, special attention was paid to such water bodies after the FDNPP accident (Konoplev et al., 2018; Tsukada et al., 2017; Wada et al., 2019; Wakiyama et al., 2017; Yoshimura et al., 2014).

Within Fukushima Prefecture, there are more than 3700 ponds of varving sizes, many of which are used for paddy water supply (Tsukada et al., 2017). Such irrigation ponds have been created over centuries in Japan for rice cultivation. Following the accident, these ponds are a concern because they are used for fishing and irrigation of agricultural fields and may cause crop contamination (Fukushima and Arai, 2014; Tsukada et al., 2017; Wada et al., 2019; Yoshikawa et al., 2014). The objective of present research was to study mid-term dynamics (2015-2019) of dissolved and particulate ¹³⁷Cs and its sediment-water distribution in three selected heavily contaminated ponds of the FDNPP exclusion zone and to elucidate basic mechanisms predetermining radiocesium mobility and bioavailability in closed and semi-closed ponds. A comparison was also made with the radiocesium dynamics and sediment-water distribution in water bodies of the same type in the areas contaminated as a result of the Chernobyl NPP accident.

2. Materials and methods

2.1. Study sites

A total of 137 small- and medium-sized ponds are located in Okuma Town in the FDNPP exclusion zone (Konoplev et al., 2018). Three of these ponds, Inkyozaka, Suzuuchi, and Funasawa were selected for the present study (Fig. 2). The selected ponds represent the main pond types in the exclusion zone and are characterized by ¹³⁷Cs deposition of 2.1–6.4 MBq/m² (Table 1) (Konoplev et al., 2018; Wakiyama et al., 2017). Inkyozaka, the nearest pond to FDNPP, is a closed pond with neither inflow nor outflow and is exclusively groundwater-fed. Funasawa is a recreational urban pond with regulated inflow and outflow, and Suzuuchi is an agricultural irrigation pond with a temporary inflow creek and no outflow.

2.2. Sampling

Raw water from the ponds was sampled once a month in 2015–2017 and at least once every two months in 2018–2019. Approximately 4 L water samples were collected from the surface layer of about half a meter depth and then filtered through 0.45 µm membrane filters (Merck Millipore Ltd, Germany) in the laboratory. Suspended sediments were collected on a filter, and the filtrate was allowed to flow through two cartridges of ANFEZH (a commercial sorbent based on cellulose that is impregnated with iron hexacyanoferrate (EKSORB Ltd., Russia)) to separate and immobilize dissolved radiocesium (Remez et al., 1998, 2016). More than 98% of dissolved ¹³⁷Cs in each water sample was captured by the first cartridge. The total activity of dissolved ¹³⁷Cs in a sample A₀ was calculated as follows: $A_0 = \frac{A_1^2}{A_1 - A_2}$, where A₁ and A₂ are the ¹³⁷Cs activities captured by the first and the second ANFEZH cartridges, respectively. Water temperature and conductivity were determined *in situ* using a multi-parameter water quality checker U-50, Horiba, Ltd., Japan.

Additionally, suspended sediments from the ponds were captured with integrating traps using the methodology of Antsiferov and Kosyan (1986). Each integrating sediment trap was a plastic cylinder 75 mm in diameter and 100 mm in height with a close-fitting lid. Around the circumference of the lid were six equally spaced holes of 7 mm diameter. There was a central sleeve in each trap to accommodate the rod on which six of these traps were mounted to form a pillar. This allowed collection of suspended

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Fig. 2. Locations of the ponds studied in the territory of Okuma Town in the vicinity of the Fukushima Dai-ichi nuclear power plant. A – Japan, B – vicinity of the FDNPP with isolines of ¹³⁷Cs deposition as of October 16, 2018 (https://ramap.jmc.or.jp/map/eng/).

Table 1

Characteristics of the studied ponds of Okuma town in the FDNPP exclusion zone.

Pond	n	Inkyozaka	Suzuuchi	Funasawa
Coordinates		37°25.499'N 141°01.05'E	37°24.950'N 140°58.791'E	37°24.363'N 140°59.173'E
Distance to FDNPP fence, km		0.24	3.75	3.50
Pond type		Closed,	Semi-closed,	Open,
		filled out	irrigation	recreational
¹³⁷ Cs deposition ^a ,		2.1 ± 1.1	6.4 ± 2.2	2.9 ± 0.9
MBq/m ²		(n = 9)	(n = 8)	(n = 7)
Water surface area, m ²		6500	4100	10,700
Catchment area, km ²		0.057	0.432	1.720
Maximum depth, m		2.0	1.0	2.5
Catchment soil type		Fluviosol	Fluviosol	Terrestrial regosol
Water quality				
K ⁺ in water ^(b) , μEq/L	42	28 ± 8	63 ± 17	49 ± 11
NH4 in water ^(c) , μEq/L	42	2.3	5.5	4.7
		(0-11)	(0-34)	(0-50)
Stable ¹³³ Cs ⁺ in water, nEq/L	39	0.13 ± 0.05	0.07 ± 0.04	0.09 ± 0.04
pH*	33	7.4 ± 0.9	7.0 ± 0.6	7.2 ± 0.4
Conductivity, µS/cm	33	96 ± 54	142 ± 24	154 ± 33
DOC*, mg/L	10	3.3 ± 0.3	4.5 ± 1.1	3.3 ± 0.7
Water temperature ^(b) , °C	33	17.5	17.9	16.3
		2.6–30.7	3.6-31.0	2.9–27.0

^a Wakiyama et al. (2017); ^(b)Mean ± standard deviation; ^(c)Above: arithmetic mean, below: range.

sediments at 6 different depths. The rod was securely fixed on a support to ensure the structure was robust enough to withstand flowing water and to preclude movement and vibrations. The trapping of suspended sediments occurred in the Suzuuchi pond from May 23, 2018 to July 13, 2018, and in the Inkyozaka and Funasawa ponds from July 10, 2019 to August 5, 2019. Applicability of these integrating sediment traps for representative sampling of all basic size fractions of sediments at relatively low currents (<10 cm/s) was documented earlier (Galas et al., 2006). After collection of suspended sediments the traps were brought to the laboratory, where turbid water was dried at 105 °C. The dried suspended material was disaggregated in a mortar and then served for radiocesium measurements. Using integrating traps, a sufficient amount of suspended sediments (more than 1 g) was collected in each of the three ponds.

Methods for sampling bottom sediments and catchment soils were described elsewhere (Konoplev et al., 2016; Wakiyama et al., 2017).

2.3. Sample analysis

The activity concentrations of ¹³⁴Cs and ¹³⁷Cs were determined in suspended material, as well as in ANFEZH sorbents. This was done based on gamma spectrometry using a GC3018 high-purity germanium detector (HPGe) by MIRION Technologies (CAN-BERRA), Inc., USA.

The concentrations of the major cations competitive with radiocesium for selective sorption sites (namely, K^+ and NH_4^+) were determined by ion chromatography using a Thermo Scientific (USA) DIONEX 1100. The stable ¹³³Cs⁺ was determined by inductively

coupled plasma spectrometer (ICP-MS) ELAN DRC 2 by Perkin Elmer, Inc., USA. The samples were prepared by adding HNO₃ to a volumetric concentration of 5%.

Dissolved organic carbon (DOC) concentration was determined using a total organic carbon analyzer TOC-L CSH, Shimadzu, CO., Japan. All samples for DOC measurements were pretreated with a 1 M HCl to remove inorganic carbon. DOC concentrations were quantified based on calibration curves made using a potassium hydrogen phthalate solution at concentrations ranging from 0.3 to 3 mgC/L.

The particle size distribution of suspended material accumulated by the sediment traps was analyzed using a laser diffraction particle size analyzer Mastersizer 3000 (Malvern Instruments Ltd., UK). The particles ranging from 0.05 μ m to 3 mm were scanned and classified into 51 categories. A portion of each dried suspended material was set up into the Mastersizer after 1–2 min of ultrasonic treatment, and each sample was scanned 5 times. The PSD curves were derived from averaged values of volume ratio in each particle size range.

Sediment and soil samples were crushed in an agate mortar and then passed through a 63 μ m mesh sieve for X-ray diffraction (XRD) and X-ray fluorescence (XRF) analyses. The clay minerals were identified using a X-ray powder diffractometer Smartlab (Rigaku, Japan). The particles were step-scanned from 5 to 80° in steps of 0.04° with Cu Kα radiation produced at 40 kV and 40 mA. XRD peak patterns were analyzed with the International Centre for Diffraction Data database to identify minerals in the sediments and soils.

Major elements of sediments and soils were analyzed using a wavelength dispersive X-ray fluorescence spectrometer RIX-1000 (Rigaku, Japan). Rh-K α radiation at 50 kV at 50 mA was used. The fundamental parameter method was used to quantify each element.

In addition, total organic carbon content in sediments and soils were measured. Samples were crushed in a mortar and then analyzed using TOC-L with solid sample measurement system SSM-5000A (SHIMADZU, Japan).

All measurements were performed in the analytical laboratory of the Institute of Environmental Radioactivity at Fukushima University.

3. Results and discussion

The ¹³⁷Cs activity concentrations in suspended matter determined from surface raw water and integrating sediment traps were similar in all the ponds during the survey period (Table 2). These results suggest that the suspended sediments were uniformly distributed across the depths in the ponds and that sampling water from the ponds' surface layers was representative of the whole water column (the correlation coefficients r were: 0.97, 0.99 and 0.98 for Inkyozaka, Suzuuchi and Funasawa ponds, respectively ($p \le 0.001$)).

The studied ponds integrate solid material delivered by surface runoff from their catchments, and by resuspension from the

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bottom. Fig. 3 shows the PSD of the suspended sediments collected using the integrating sediment traps from the ponds. The PSD of suspended sediments from all three ponds showed two slight peaks. The first one for all three ponds was at approximately 1 μ m (clay), whereas the second slight peak was observed at 430 μ m (medium sand) in Inkyozaka, at 920 μ m (coarse sand) in Funasawa, and at 2000 μ m (coarse sand) in Suzuuchi. Sediments from Suzuuchi and Funasawa had one major peak at approximately 30 μ m (coarse silt), whereas sediments from Inkyozaka had two major peaks at approximately 3–5 μ m (fine silt) and 50 μ m (coarse silt), respectively. For all studied ponds, accumulated suspended material is mainly presented by clay and silt fractions. This corresponds to the results of a recent study (Komissarov and Ogura, 2020) which has shown that eroded from a catchment fine particles are dominant in lake bottom sediments.

The typical XRD peak patterns of soil and sediment particles confirm the presence of micaceous clay minerals, which selectively adsorb and fix radiocesium, in sediments and catchment soils of the Suzuuchi pond (Fig. 4a). Similar peak patterns were observed for soils and sediments from other ponds as well. Fig. 4b shows vertical distributions of ¹³⁷Cs in three individual cores of bottom sediments from the Suzuuchi pond and demonstrates that maximal activity concentrations were observed in the top layer of sediments at least up to the summer 2016 when the cores were collected. Major elements are distributed rather uniformly in bottom sediments (Fig. 4c). TOC in the top layer of bottom sediments varied in relatively wide range – from 4% to 12% depending on location. The lowest TOC was observed at the inlet of the temporal creek to the Suzuuchi pond where coarse sediments deposited.

Fig. 5 shows the time dependence of the mean annual ¹³⁷Cs activity concentrations from 2015 to 2019 in the studied ponds. In all three ponds, the dissolved and particulate ¹³⁷Cs activity



Fig. 3. Differential particle size distribution (PSD) of the suspended sediments collected using the integrating traps exposed in the vertical cross section of the ponds.

Table 2

¹³⁷Cs activity concentrations in the suspended sediments (SS) collected with the integrated traps and SS extracted by the filtration of 4 L surface layer water through 0.45 μm filter for ponds under study.

Pond	Inkyozaka	Suzuuchi	Funasawa
¹³⁷ Cs in SS trapped, Bq/kg	$(1.3\pm 0.02)\times 10^{5(a)}$	$(1.1 \pm 0.3) imes 10^{5(b)}$	$(0.78 \pm 0.02) \times 10^{5(a)}$
Period of SS trapping	July 10, 2019 –	May 23, 2018 —	July 10, 2019 –
	August 5, 2019	July 13, 2018	August 5, 2019
Average ¹³⁷ Cs in SS filtered, $Bq/kg^{(c)}$ (n = 4)	$(1.0 \pm 0.2) imes 10^5$	$(1.0 \pm 0.4) imes 10^5$	$(0.77 \pm 0.19) imes 10^5$

^(a)Gamma spectrometry measurement error of individual trapped SS sample. ^(b)Arithmetic mean with the standard deviation for six separate traps ^(c)Arithmetic mean with the standard deviation for surface water sample measurements around the time of SS trapping.

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Fig. 4. Typical characteristics of sediment samples from Suzuuchi pond collected in July 2016, (a) X-ray diffraction pattern of top layer of the bottom sediments and soil sample from the pond's catchment (V – vermiculite, B – biotite, A – amphibole, K - kaolinite; (b) vertical distribution of ¹³⁷Cs in three cores of bottom sediments BS-1, BS-2 and BS-3 (Wakiyama et al., 2017); (c) vertical distribution of major elements in the core of bottom sediments BS-1; (d) vertical distribution of TOC in three bottom sediment cores BS-1, BS-2 and BS-3.

concentrations decreased: for Inkyozaka $[^{137}Cs]_p$ – from $(3.5\pm1.7)\times10^5$ Bq/kg in 2015 to $(1.1\pm0.2)\times10^5$ Bq/kg in 2019, $[^{137}Cs]_d$ – from 1.6 ± 0.2 Bq/L in 2015 to 1.3 ± 0.3 Bq/L in 2019; for Suzuuchi $[^{137}Cs]_p$ – from $(3.6\pm1.1)\times10^5$ Bq/kg in 2015 to $(0.6\pm0.1)\times10^5$ Bq/kg in 2019, $[^{137}Cs]_d$ – from 3.5 ± 1.4 Bq/L in 2015 to $(0.9\pm0.4$ Bq/L in 2019; for Funasawa $[^{137}Cs]_p$ – from $(3.2\pm1.3)\times10^5$ Bq/kg in 2015 to $(1.1\pm0.5)\times10^5$ Bq/kg in 2019, $[^{137}Cs]_d$ – from 1.7 ± 0.6 Bq/L in 2015 to 1.3 ± 0.9 Bq/L in 2019. The quantitative characteristics of ^{137}Cs decline in the Okuma ponds are presented in Table 3. Herein, λ_{eff} is the effective rate constant for the decline in the ^{137}Cs activity concentration; λ_{env} is the ecological rate constant for the decline in the ^{137}Cs decay equal to 0.023 year $^{-1}$; and τ_{env} is the timescale of the ^{137}Cs decline due to natural attenuation processes. It follows from Fig. 5 and Table 3 that the particulate ^{137}Cs for all three ponds during 2015–2019.

Table 4 presents data on the ¹³⁷Cs exchangeability in the surface (top 3 cm) layer of sediment in the three ponds in comparison with similar soil data for the surrounding catchment. The exchangeable fraction was extracted by 1 M ammonium acetate solution (pH = 7) at solid-liquid ratio 1:10 (Wakiyama et al., 2019) using the conventional methodology of sequential extractions (Pavlotskaya, 1974; Bobovnikova et al., 1991; Oughton et al., 1992; Tsukada et al., 2008). The soils were characterized by quite low fractions of exchangeable ¹³⁷Cs, including when compared with the Chernobyl values (Bobovnikova et al., 1991; Konoplev, 2020; Konoplev et al., 1988), whereas the surface sediment layer, formed as a result of sedimentation, is characterized by a relatively high fraction of exchangeable ¹³⁷Cs (up to 40% in Inkyozaka). Using the data

of the particulate and dissolved ¹³⁷Cs in the ponds, and the ¹³⁷Cs exchangeable fraction in the surface layer of bottom-sediments, we calculated the apparent distribution coefficient (K_d^{ex}) in the Okuma ponds, and the radiocesium interception potential RIP^{ex} using equation (5) (Table 4). Generally speaking, equation (5) could be expanded with a term accounting for stable cesium, considering that stable cesium (¹³³Cs) is characterized by higher selectivity for FES than potassium and ammonium: $K_c^{FES}(Cs/K) \approx 1000$ (Wauters et al., 1996a). However, the stable Cs concentrations in the ponds are more than five orders of magnitude lower than the concentrations of potassium and ammonium (Konoplev et al., 2018; Wakiyama et al., 2017), and therefore, the contribution of the naturally occurring stable Cs to the ¹³⁷Cs desorption of FES can be ignored compared to potassium and ammonium.

The obtained estimates of $RIP^{ex}(K)$ were relatively high, ranging from 1650 mEq/kg for Inkyozaka to 2250 mEq/kg for Suzuuchi and Funasawa. This demonstrates the ability of sediment particles to selectively adsorb ¹³⁷Cs, the results being consistent with the values measured in laboratory experiments (Nakao et al., 2014, 2015a; Yamaguchi et al., 2017) for soils typical of the contaminated areas around Fukushima. The ability of the studied sediments to selectively adsorb radiocesium is even a bit higher than for sediments in Lake Constance in Europe (Konoplev et al., 2002), which are characterized by high affinity to Cs. For all three ponds, a linear trend was obtained for the dependences of K_d^{ex} (L/kg) on ($[K^+]+5[NH_4^+])^{-1}$ (L/mEq), with the slopes conforming to calculated values $RIP^{ex}(K)$ (Table 4).

In the areas contaminated by the Chernobyl accident, the values of the radiocesium distribution coefficient in the closed lakes and

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Fig. 5. Time dependence of the particulate and dissolved ¹³⁷Cs activity concentrations for 2015 to 2019 in the ponds of Okuma Town in the vicinity of Fukushima Dai-ichi nuclear power plant (FDNPP): (a) Inkyozaka, (b) Suzuuchi, and (c) Funasawa. λ_{eff}^p and λ_{eff}^d are the first order rate constants of the particulate and dissolved ¹³⁷Cs activity concentrations decline in the pond, respectively. Number of water samples per year: n(2015) = 5; n(2016) = 12; n(2017) = 12; n(2018) = 6; n(2019) = 9.

Table 3

Kinetic characteristics for the time dependence of ¹³⁷Cs activity concentrations in the studied ponds of Okuma town: $\lambda_{eff}^p(yr^{-1})$ and $\lambda_{eff}^d(yr^{-1})$ are the effective rate constants of particulate and dissolved ¹³⁷Cs activity concentration's decline, respectively; $\lambda_{env}^p(yr^{-1})$ and $\lambda_{env}^d(yr^{-1})$ are the ecological rate constants of the particulate and dissolved ¹³⁷Cs activity concentration's decline, respectively; $\tau_{env}^p(yr)$ are the time scales of respective processes.

	Particulate ¹³⁷ Cs			Dissolved ¹³⁷ Cs		
	λ_{eff}^p , yr ⁻¹	λ_{env}^p , yr ⁻¹	$ au_{env}^p$, yr	λ_{eff}^d , yr ⁻¹	λ_{env}^d , yr ⁻¹	$ au^d_{\mathit{env}}$, yr
Inkyozaka Suzuuchi	0.254	0.231	4.3	0.085	0.062	16.1
Funasawa	0.327	0.304	3.3	0.280	0.203	13.9

ponds was much lower than those in the rivers and large open lakes (Konoplev et al., 1992a,b; Konoplev et al., 1998, 2002). For example, in the closed Lake Svyatoe in the Bryansk region, Russia (about 250 km NE of the Chernobyl NPP) in 1993–1995 (7–9 years after the Chernobyl accident), the apparent K_d was $(3.5 \pm 0.7) \times 10^3$ L/kg (Konoplev et al., 1998). Similar values of K_d (8.1 × 10³ L/kg) were reported for the closed organic Lake Vorsee in Baden-Wurttemberg, Germany (about 2000 km west of the Chernobyl NPP) over the same period (Konoplev et al., 2002). In contrast, in the large open Lake Constance (Germany) having a high self-purification capacity with respect to radiocesium, the $K_d(^{137}Cs)$ values were $(1-10) \times 10^5$ L/kg (Konoplev et al., 2002), which is close to the apparent $K_d(^{137}Cs)$ in the freshwater bodies of the FDNPP contaminated areas.

The apparent K_d (¹³⁷Cs) values in the studied ponds of Okuma Town appeared to be at least an order of magnitude higher than those in the closed lakes Svyatoe and Vorsee and close to the values in Lake Constance (Konoplev et al., 1998, 2002). They are also similar to those in the rivers and dam reservoirs of the FDNPP contaminated area (Konoplev, 2015; Yoshimura et al., 2015). This can be attributed to two factors. The first is the relatively high binding ability of the soils and sediments in the Fukushima contaminated area (Nakao et al., 2015a; Yamaguchi et al., 2017), and the second factor is occurrence of insoluble glassy hot particles in the Fukushima fallout and their higher content in the FDNPP near zone (Ikehara et al., 2018; Satou et al., 2018).

Fig. 6 presents the time dependence of the ¹³⁷Cs apparent distribution coefficients in the ponds based on the 2015–2019 observations. A faster reduction in the particulate compared to dissolved ¹³⁷Cs concentration in the ponds (Fig. 5) was expected to lead to a decrease in the K_d value for this time period, which is corroborated by Fig. 5. The rate constants of K_d decline for the ponds are in the range 0.12–0.18 yr⁻¹.

The trend of decline in the apparent distribution coefficient K_d revealed for all three ponds (Fig. 6) could be associated with ¹³⁷Cs leaching from glassy CsMP (Konoplev et al., 2018) which can contribute up to 40% and more of the deposition inventory in the FDNPP near zone (Ikehara et al., 2018, 2020).

The process of decomposition of CsMP and associated leaching out of ¹³⁷Cs seems to be very slow. It is thought to be even slower than for Chernobyl-derived fuel particles. The point is that the UO₂ of fuel particles is oxidized by atmospheric oxygen resulting in the formation of UO₃ which is much more water soluble than UO₂. This process accelerates decomposition of fuel particles and radionuclides leaching from them (Beresford et al., 2016; Konoplev, 2020). Silicates comprising glassy hot particles are not oxidized in the environment and are expected to be more persistent in soils and sediments than uranium oxides. The CsMP decomposition is also much slower than the ¹³⁷Cs fixation by micaceous clay minerals, the rate constant of which is $k_f = 4.4-5.5 \text{ yr}^{-1}$ (Konoplev et al., 1992a, 1996). Therefore, 137 Cs leaching from CsMP is a limiting factor in the process of 137 Cs chemical form transformation in the sediment-water system (Fig. 1). This means that the rate constant of K_d 's decline can serve at least as an upper estimate for the rate constant of ¹³⁷Cs leaching from CsMP (Fig. 1). In reality, the process can be even slower since other geochemical and biological processes, such as stagnation and eutrophication, can also contribute in ¹³⁷Cs remobilization. We estimated the first order rate constants of $^{137}\mbox{Cs}$ leaching from $\mbox{CsMP}\left(k_l\right)$ based on the results of laboratory experiments on ¹³⁷Cs dissolution from individual particles in pure and sea waters at 30 °C (Okumura et al., 2019) and obtained the values 0.14 \pm 0.01 year⁻¹ for pure (deionized) water and 2.1 \pm 0.4 year⁻¹ for seawater. Extrapolating temperature dependence of k_1 derived from laboratory experiments of Okumura et al. (2019) for the range 30 °C–90 °C to the mean annual water temperature in the

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Table 4

Characteristics of the137Cs s	speciation, distribut	ion and exchangeable	e interception p	potential in the sedir	ment-water system of	the Okuma ponds.
	•					

Pond	Inkyozaka	Suzuuchi	Funasawa
Bottom sediments (3 cm deep surface layer) ¹³⁷ Cs exchangeable fraction ^a , $%$ (n = 3) Soils (2 cm deep surface layer single measurements)	44 ± 5	22.6 ± 2.3	19.6 ± 4.4
137 Cs exchangeable fraction ^a , %	3.6	$\begin{array}{c} 0.4 \\ 1.2 \times 10^5 \ (0.6 - 2.3) \times 10^5 \\ 2.2 \times 10^4 \ (-1.5 \ 2.3) \times 10^4 \end{array}$	3.0
$K_d^{(b)}$, L/kg	$1.4 \times 10^5 (0.4 - 3.5) \times 10^5$		1.6 × 10 ⁵ (0.3–3.8) × 10 ⁵
$K_d^{e_X(U)}$, L/kg	$5.5 \times 10^{4} (0.2 - 1.5) \times 10^{3}$	$2.9 \times 10^{4} (1.4 - 5.2) \times 10^{4}$	$3.3 \times 10^4 (0.8 - 7.5) \times 10^4$
RIP ^{ex} (K), meq/kg	1650	2250	2250

^a Extracted by 1 M CH₃COONH₄ solution at solid-liquid ratio 1:10 (Wakiyama et al., 2019); ^(b)Above – arithmetic mean, below – range.



Fig. 6. Time dependence of the ¹³⁷Cs apparent distribution coefficient K_d for the suspended sediment–water system in the Okuma Town ponds of the FDNPP exclusion zone; k is the first order rate constant of sediment-water apparent K_d decline: (a) Inkyozaka, (b) Suzuuchi, and (c) Funasawa.

ponds – about 17 °C (Table 1), we get $k_1 = 0.044 \text{ yr}^{-1}$ for pure water and $k_1 = 0.44 \text{ yr}^{-1}$ for seawater. It would be reasonable to assume that the rate constant of ¹³⁷Cs leaching from CsMP in sedimentwater system of real ponds in the Fukushima exclusion zone could be in between these two values since the conditions there are more chemically and biologically aggressive than in the batch with pure water but less aggressive than in seawater. This consideration supports our hypothesis that the observed unusual reduction of apparent K_d (¹³⁷Cs) in the Okuma ponds in 2015–2019 may be associated with CsMP decomposition and subsequent ¹³⁷Cs remobilization. At the same, time there could be alternative mechanisms of ¹³⁷Cs remobilization in ponds such as their eutrophication and stabilization of the frayed edges of clay mineral's interlayers by organic matter and inhibition of ¹³⁷Cs fixation (Beresford et al., 2016; Tachi et al., 2020), thus this phenomenon requires further investigation.

Seasonal variations in dissolved ¹³⁷Cs concentrations in the studied ponds of the FDNPP exclusion zone for 2016-2017 are shown in Fig. 7. The ¹³⁷Cs concentrations generally increased significantly in the summer-early autumn. In the Inkyozaka and Funasawa ponds, the highest concentrations were observed from July to October, whereas in the Suzuuchi pond the highest concentration of ¹³⁷Cs occurred in June. Similar seasonal variations of dissolved ¹³⁷Cs were observed for the rivers in the contaminated area (Nakanishi and Sakuma, 2019; Tsuji et al., 2016). Nakanishi and Sakuma (2019) related these seasonal variations to the changes in the river water temperature and the leaching of dissolved ¹³⁷Cs from decomposing litter in flooded areas during the summer season. Conceivably, the changes in water temperature could have an impact on ion-exchange desorption of ¹³⁷Cs which proceeds according to the Gibbs-Helmholz and/or Arrhenius equations (Lee et al., 2017; Liu et al., 2003). Assuming that the proportion of exchangeable ¹³⁷Cs in sediments does not change much over time and the filtration of samples at room temperature does not lead to a significant redistribution of ¹³⁷Cs between the solid and liquid phases, we can roughly estimate the ¹³⁷Cs desorption activation energy (E_a) for the sediments from the studied ponds. E_a can be derived from the slope of the linear relationship between $\ln(K_d^{ex})$ and the reverse absolute temperature (degrees K). The estimates we obtained were 16 kJ/mol for Suzuuchi and 22 kJ/mol for Inkyozaka respectively; this data agrees well with the results of laboratory experiments determining E_a for selective sorption sites of micaceous clay minerals (Liu et al., 2003).

Similar seasonal variations of dissolved ¹³⁷Cs activity concentration were also observed in the Chernobyl Cooling Pond (Kanivets et al., 2020), and these were attributed to the variations in ammonium concentration in the pond's bottom sediments pore water. In the studied Fukushima ponds, the decomposition of organic matter in the reducing conditions of bottom-sediments could also have led to ammonium generation in pore water, which could have contributed in some extent to increased dissolved ¹³⁷Cs in the pond's water column in summer, thus this



Fig. 7. Seasonal variations of the dissolved ¹³⁷Cs activity concentrations in the water of the Okuma Town ponds in the FDNPP exclusion zone: (a) Inkyozaka, (b) Suzuuchi and (c) Funasawa.

should be further studied in the future.

4. Conclusions

The studies of ¹³⁷Cs behavior in the ponds of Okuma Town in the vicinity of FDNPP conducted during 2015–2019 have shown that the concentrations of this radionuclide are higher in ponds than in rivers and dam reservoirs of the region. The highest levels of the dissolved ¹³⁷Cs in the studied ponds were observed from June to October, depending on the pond and monitoring year. Additional dissolution of ¹³⁷Cs in the summer can be attributed to temperature dependence of ¹³⁷Cs desorption from FES and its remobilization by

ammonium.

The apparent $K_d(^{137}Cs)$ in the studied ponds, in absolute value, appeared to be much higher than that for closed lakes of the Chernobyl contaminated area; however, these values were comparable to those characteristic of the rivers and reservoirs of the FDNPP contaminated area. Most likely, the elevated values of $K_d(^{137}Cs)$ could be explained by two major reasons: (1) high values of binding ability of soils and sediments in Fukushima contaminated areas in relation to radiocesium, and (2) the presence of water-insoluble CsMPs in the Fukushima accidental fallout. The ¹³⁷Cs exchangeable distribution coefficients K_d^{ex} for all three ponds were found to be linearly proportional to the inverse effective concentration of major Cs competitive cations $([K^+] + 5[NH_4^+])^{-1}$, as would be expected from the theory of radiocesium selective sorption on the FES of micaceous clay minerals.

Based on data about the ¹³⁷Cs speciation at the surface layer of the bottom-sediments in the three ponds and its distribution in the sediment–water system, the exchangeable $RIP^{ex}(K)$ was estimated to be 1650–2250 mEq/kg, which is within the range of values measured in the laboratory and available in literature.

Despite ¹³⁷Cs contamination in the studied ponds being relatively persistent, a decline in both particulate and dissolved ¹³⁷Cs activity concentrations was revealed. The reduction rate of particulate ¹³⁷Cs activity concentrations was much higher than that for dissolved ¹³⁷Cs. Thus, the apparent distribution coefficient K_d (¹³⁷Cs) in the sediment–water system decreased with the rate constant 0.12–0.18 yr⁻¹. We hypothesize that the K_d (¹³⁷Cs) decline may be associated with the decomposition of CsMP and subsequent ¹³⁷Cs leaching from them, since the rate of K_d decline in the ponds corresponds to the roughly estimated rate of ¹³⁷Cs leaching from CsMP in freshwater environment based on the latest available data of laboratory experiments.

Monitoring of changes in radioactive contamination of the ponds in the FDNPP exclusion zone is essential not only in the context of radiation safety, but also for better understanding of main mechanisms responsible for persistence, mobility and bioavailability of Fukushima-derived radiocesium in the sedimentwater system. These ponds are valuable and convenient subjects for study of the surrounding freshwater environment with all its features and complexities.

Study of distribution and dynamics of radionuclides in real-life water bodies makes necessary using assumptions and hypotheses to explain what we observe. Such assumptions and models are sources of uncertainties that can be quite high. Even when interpretation seems to be valid, it still requires confirmation. Thus, laboratory experiments on particular mechanisms and processes occurring in real objects can be used to corroborate hypotheses and complement obtained field data. Another way of ascertaining results is continuation of observations which can either confirm our assumptions and models, or prove them wrong. To this end, the presented monitoring efforts should be maintained in future.

Credit author statement

Alexei Konoplev: Conceptualization, Writing – original draft preparation; Yoshifumi Wakiyama: Investigation, Data curation; Toshihiro Wada: Writing -Review & editing; Cameron Udy: Investigation, Writing -Review & editing; Volodymyr Kanivets: Methodology, Investigation; Maxim Ivanov: Investigation, Visualization; Mikhail Komissarov: Investigation; Tsugiko Takase: Methodology, Data curation; Azusa Goto: Resources; Kenji Nanba: Supervision.

Declaration of competing interest

The authors declare that they have no competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research was partially supported by the Japan Society for the Promotion of Science, Grant-in-aid for Scientific Research (B) (KAKENHI 18H03389) and by the Science and Technology Research Partnership for Sustainable Development, the Japan Science and Technology Agency/Japan International Cooperation Agency (JPMJSA1603).

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