Journal of Environmental Management 320 (2022) 115905

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Research article

Radiocesium accumulation in Lake Kasumigaura by riverine input and migration following the Fukushima Dai-ichi nuclear power plant accident

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ARTICLE INFO

Keywords:
Fukushima nuclear accident
Radiocesium
Sediment cores
Suspended solids
Riverine input
Lake kasumigaura

ABSTRACT

Vertical radiocesium concentration profiles and inventories in sediments were measured in Lake Kasumigaura following the 2011 Fukushima Dai-ichi Nuclear Power Plant accident. Further measurements of radiocesium concentrations in suspended solids (SS) have been conducted since September 2012 in the Koise and Sakura rivers inflowing into the lake. Cesium-137 (^{137}Cs) accumulated intensively near the inflow outlets in the lake. At the lake center, the ^{137}Cs inventory in sediments increased during 2011–2014; however, few changes were observed during 2014–2016. The ^{137}Cs surface concentration and inventory decreased considerably in Tsuchiura-iri Bay until 3 years after the accident, indicating ^{137}Cs migration. However, the rate of decrease subsequently slowed due to the ^{137}Cs supply from the river. The ^{137}Cs concentration in river SS declined during 2012–2015; however, it remained 1–2 orders of magnitude above its pre-accident level. The entrainment coefficient of particulate ^{137}Cs in the inflows was initially higher in the Koise River but decreased exponentially more rapidly in the Koise River than in the Sakura River until 2015. Therefore, in the future, the difference in ^{137}Cs concentrations will be smaller. The ^{137}Cs concentration in the Koise River will continue to decrease; thus, the difference in the ^{137}Cs inventory between the northern and southern parts of the lake will decrease. Total estimated amounts of ^{137}Cs in the entire lake were 3.72×10^{12} Bq in December 2012 and 4.18×10^{12} Bq in August 2016. The accumulated amount of ^{137}Cs in the entire lake based on sediment analysis was similar to the riverine input of particulate ^{137}Cs based on riverine SS analysis from December 2012−;August 2016, confirming the high trapping performance of the lake for particulate matter provided by the basin. Moreover, the amount of ^{137}Cs accumulated in the lake in 2016 may have originated from comparable rates of atmospheric deposition and riverine input. These findings provide useful insights for future prediction and management of radiocesium contamination and the effects of riverine inputs in general shallow lakes.

1. Introduction

For sustainable management and use of water resources, it is essential to address water pollution issues. Water pollution includes various organic and inorganic water pollutants, such as the drugs and pharmaceutical residues (Basheer, 2018; Ali et al., 2018; Basheer and Ali, 2018), as well as radioactive contamination from nuclear power plant accidents, are becoming a subject of global anxiety. The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, following a massive earthquake and tsunami disaster on Mar 11, 2011, resulted in the substantial release of radionuclides and widespread radioactive contamination of surrounding areas, mainly from 15–16 March 2011 (Hirose, 2012). In particular, Cesium-137 (^{137}Cs) remains in the environment because of its long half-life (30.1 years). A significant portion of the ^{137}Cs deposited on the ground firmly adheres to the soil (Qin et al., 2012), and subsequent transfer of the deposited ^{137}Cs through river systems is reported (e.g., Onda et al., 2020; Feng et al., 2022). Therefore, understanding the dynamics of ^{137}Cs in downstream lakes is essential from the viewpoint of early-stage management and prediction of the fate of radioactive fallout in the environment.

After the fallout of radionuclides, a specific portion of the fallout is deposited into lakes (Davison et al., 1993; Yoshimura et al., 2014). Subsequently, ^{137}Cs is transported into lakes through inflow from rivers and deposited to the surface of the bottom sediment (Onda et al., 2020).

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https://doi.org/10.1016/j.jenvman.2022.115905
Received 11 May 2022; Received in revised form 19 July 2022; Accepted 28 July 2022
Available online 11 August 2022

Several studies have assessed the contribution of ^{137}Cs fluvial discharge to lakes by investigating the amount accumulated in bottom sediments relative to deposition around the lakes (He et al., 1996; Ilus and Saxén, 2005; Matsunaga et al., 1999). To date, few studies have investigated the riverine load and flux of ^{137}Cs flowing into the lake and compared them with the change in the ^{137}Cs inventory of the lake sediment. This information is crucial for determining the amount of atmospheric fallout and riverine input to evaluate the lake environment.

Lake Kasumigaura is the second largest lake in Japan. Due to the closure of the gate at the effluent river, the lake has a relatively long water retention time; therefore, it may be a terminal location of ^{137}Cs, which is deposited in the catchment area. Several studies have investigated the ^{137}Cs distribution and inventory in lake sediments. Fukushima et al. (2010) collected sediment cores at the center of Lake Kasumigaura and estimated the mass sedimentation rate by analyzing the ^{137}Cs peak derived from atmospheric nuclear tests. After the FDNPP accident, Fukushima et al. (2018) observed the rapid penetration of ^{137}Cs in sediments at the lake's center. They suggested that wind-induced stress and sediment porosity are key parameters. Tsuji et al. (2019) investigated the spatial distribution of ^{137}Cs in the lake sediments in the 3 years following the FDNPP accident; however, the flux from the riverine input based on a continuous survey was not shown. To the best of our knowledge, to date, there have been no discussions regarding the ^{137}Cs concentrations in both riverine suspended solids (SS) and lake sediments after 3 years of fallout. The ^{137}Cs contamination of lake sediments regulates the ^{137}Cs concentration in lake water and organisms, such as demersal fish (Fukushima and Arai, 2014; Matsuzaki et al., 2021). Therefore, the vertical and horizontal transfer of ^{137}Cs in lake sediments must be monitored widely and continuously to predict the future conditions of Lake Kasumigaura.

The objectives of this study were three-fold: (1) to investigate the temporal/spatial changes in inflow and accumulation of particulate ^{137}Cs in Lake Kasumigaura for 5 or 6 years following the FDNPP accident; (2) to discuss the factors explaining the observed trends, such as a yearly decrease in ^{137}Cs concentrations (in riverine SS and lake sediments) and an increase/decrease in the ^{137}Cs inventories in lake sediments among different sites, primarily focusing on the horizontal transfer of fine particles; and (3) to calculate the riverine input and accumulation of ^{137}Cs in the lake to confirm the contribution of the riverine input to the lake sediments. The findings provide valuable information on the river contribution, post-depositional processes, and changes that may occur in the future involving radiocesium in Lake Kasumigaura.

2. Material and methods

2.1. Study area

Lake Kasumigaura is located in Ibaraki Prefecture in the eastern region of the Kanto Plain, approximately 160 km south-south-west of the FDNPP (Fig. 1). It has a surface area of 172 km^2, a mean depth of 4 m, and a maximum depth of 7 m. The mean water retention time is approximately 200 d. The Koise and Sakura rivers are the main rivers flowing into two large bays of the lake, Takahama-iri, and Tsuchiura-iri, respectively. Water flows through the lake from the northwest to the southeast and drains into the Hitachitone River. The lake is sufficiently shallow that the vertical stratification is easily destroyed by moderately strong winds. The lake changed from a brackish into a freshwater lake 5 years after the Hitachitone River gate to the Pacific Ocean was implemented in 1963 for purposes of flood defense, desalination, and water utilization.

The lake has a 1426-km^2 catchment area (excluding the lake area). The high-resolution land-use and land-cover map obtained by the Advanced Land Observing Satellite (ALOS-2; JAXA, 2017) showed that the catchment comprised 37% forests, 28% paddy fields, 23% crop fields, 7% urban area, 1% water surface, and 4% other areas (Table 1). The significant surface geology in the catchment is loam. The climate of the area is similar to that of other regions on the Pacific side of Japan, with an annual mean air temperature of approximately 14 °C and annual precipitation of 1250 mm.

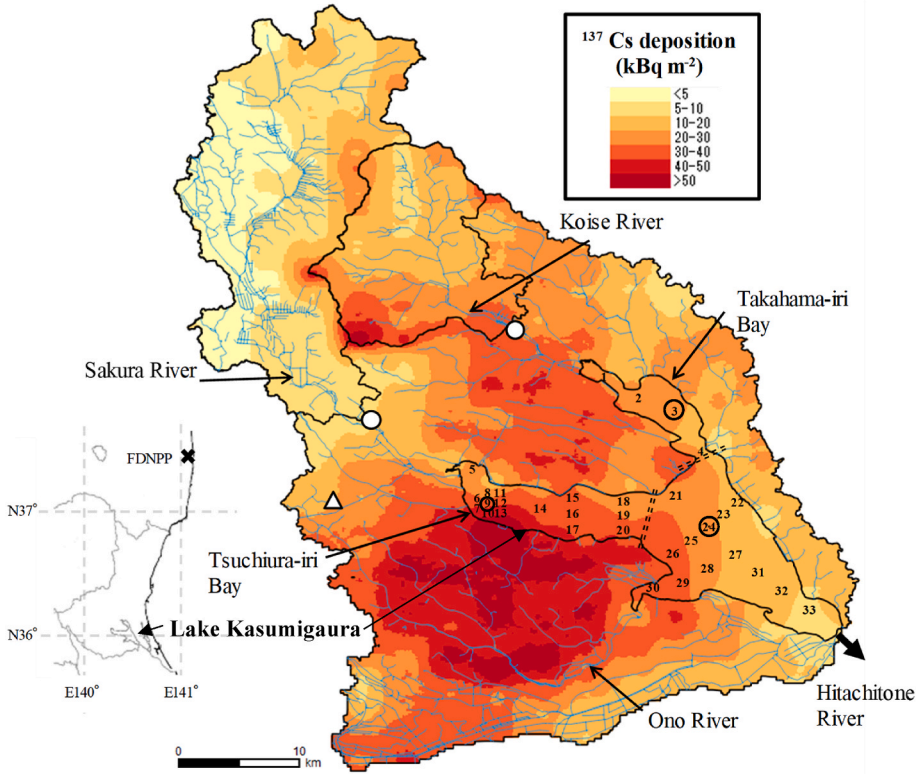

Fig. 1. Location of the study site. Numbers indicate the sediment core sampling points in Lake Kasumigaura. The circled numbers are locations where samples were taken almost every year. Open circles represent the suspended solid (SS) sampling points on Koise and Sakura rivers. Single broken lines show the watershed area at each point. The open triangle represents the observation site of the atmospheric deposition reported by Igarashi et al. (2015). FDNPP - Fukushima Dai-ichi Nuclear Power Plant.

Table 1
Summary of the research target basin.

Basin	Area (km^2)	Land cover (%)						^{137}Cs deposition (kBq m^{-2})
		Forest	Paddy	Crop	Urban	Water	Others	
Lake Kasumigaura basin[a]	1426	37	28	23	7	1	4	23.3
Koise River basin[b]	0227	65	15	15	2	0	3	21.4
Sakura River basin[b]	0353	43	27	21	5	0	4	7.8
Experimental basin (Koise + Sakura River)[b]	0580	51	22	19	4	0	4	13.1

[a] Excluding the lake.
[b] Catchment area at the sampling points.

2.2. Atmospheric radiocesium deposition in the catchment area and Lake Kasumigaura

Kato et al. (2019) reconstructed the initial fallout map of Fukushima accident-derived radiocesium in the eastern part of Japan based on an analysis of airborne monitoring survey datasets. The ^{137}Cs aerial deposition densities derived from the third, fifth, and the prefecture airborne monitoring surveys were compared and adjusted for variation in the measured radioactivity associated with the influence of multiple factors on radioactive decay. These factors included the half-life of radiocesium, natural weathering processes, variation in the calibration procedure used between the airborne monitoring surveys, and other unidentified processes, which may also have an influence. Finally, calibrated deposition densities for each land-use type from the fifth airborne monitoring survey were used to complement the fallout map derived from the third airborne monitoring survey. The data were downloaded from CRiED (2018). A value of 2.5 kBq m^{-2} was assigned in the area of ^{137}Cs deposition <5 kBq m^{-2} (under the detection limit), accounting for only 7% of the entire basin area. Radioactivity was corrected for decay to Mar 15, 2011, when the contamination of radionuclides occurred around the study area.

Deposition may have been affected by the rapid runoff events after the initial fallout. Igarashi et al. (2015) analyzed atmospheric deposition samples collected at the Meteorological Research Institute (MRI) in Tsukuba, Ibaraki (approximately 10 km west of the lake [Fig. 1]) since the 1980s. At this site, the ^{137}Cs deposition determined by airborne monitoring (23 kBq m^{-2}) was similar to the value observed by Igarashi et al. (2015) during March 2011 (23 ± 0.9 kBq m^{-2}). Therefore, we approximated the initial deposition using the airborne monitoring results. The mean deposition of ^{137}Cs in the watershed area was estimated to be 23.3 kBq m^{-2} (Fig. 1; Table 1). Deposition was <72 kBq m^{-2} throughout the entire basin. A relatively high amount of deposition was confirmed in the southwest of the basin.

The initial atmospheric deposition of ^{137}Cs on the lake surface was estimated by applying spline interpolation to the watershed deposition observed by airborne monitoring (Fig. 1). The average ^{137}Cs deposition in the lake was estimated to be 24.9 kBq m^{-2}.

2.3. Sediment core sampling in Lake Kasumigaura and laboratory analysis

Sediment cores were collected at 3, 3, 5, 3, 6, 12, and 25 sites in 2007, 2011, 2012, 2013, 2014, 2015, and 2016, respectively. Sampling details are presented in Table 2. Most core samples were collected by scuba divers using acrylic tubes with an inner diameter of 10 cm (core depth: >50 cm except for one core), and seven cores were obtained from gravity corers with acrylic tubes with an inner diameter of 7 or 11 cm (core depth: >10 cm). Minimal differences in ^{137}Cs vertical profiles existed between the cores taken by scuba divers and those taken by a gravity corer (Arai et al., 2017). Most cores were sliced at a 2-cm interval, except for the core taken at site 9 in 2011 (3-cm interval). In 2015 and 2016, 34 cores were not sliced but homogenized and sampled for the measurement of the total inventory. Water content and specific gravity were analyzed with wet and dry weights obtained by drying at 110 °C for 12–24 h using a pycnometer. The apparent density was calculated from the water content and specific gravity. Loss on ignition was analyzed with a dry and ignited weight obtained by heating at 650 °C for 3 h in a muffle furnace.

Table 2
Summary of the sediment core sampling in Lake Kasumigaura.

Sampling date (year/month/day)	Site	Method[a]	N[b]	Length (cm)	Cut interval (cm)
2007/6/19	3, 9, 24	S	1	>50	2
2011/9/7	9	G	1	15	3
2011/9/30	24, 33	G	1	10	2
2012/7/26	24	S	1	>50	2
	1, 3, 9, 16	G	1	10	2
2013/11/12	3, 9, 24	S	2	>50	2
2014/9/29 or 30	1, 3, 5, 9, 24, 33	S	1	>50	2
2015/9/17	9, 16	S	1	>50	2
	6–13, 16, 19, 24, 31	S	1	60–90	Not[c]
2016/8/24 or 25	3, 9, 24	S	1	>50	2
	1, 2, 4, 5, 14–23, 25–30, 32, 33	S	1	30–70	Not

[a] S, scuba divers using acrylic tubes; G, gravity core sampler.
[b] Number of the cores collected at each site.
[c] The 0–2 cm sediment was also analyzed at site 24.

Wet samples were freeze-dried or dried at 105 °C for 24 h and homogenized. Radioactive concentrations of ^{134}Cs and ^{137}Cs in the sediment samples were determined using gamma-ray spectroscopy. Gamma-ray emissions at energies of 604 keV (^{134}Cs) and 661 keV (^{137}Cs) were measured using a high-purity n-type germanium coaxial gamma-ray detector (EGC25-195-R, Canberra-Eurisys, USA). The measurement system was calibrated, as reported by Kato et al. (2012). Analytical accuracy was certified via a World-Wide Proficiency Test (IAEA, 2007) using standard soil samples from International Atomic Energy Agency (IAEA). The counting error in measurements was <10%. Radioactive concentrations were corrected for decay on Mar 15, 2011. The ^{137}Cs inventories (Bq m^{-2}) were computed based on the ^{137}Cs concentration (Bq kg^{-1} of dried sediments), apparent density (kg m^{-3}), and sediment depth (m). For a few sediment samples taken from a depth of 0–2 cm in September 2014, the median diameters of the sediment particles were measured using a laser diffraction particle size analyzer (SALD-3100, Shimadzu Co., Ltd., Kyoto, Japan) after H_2O_2 digestion. Size ranges from 0.05 to 3000 μm were scanned and classified into 51 categories.

2.4. SS sampling in the two main inflows and laboratory analysis

SS was collected approximately monthly from September 2012−; December 2015 and from October−;November 2009 (prior to the accident) in the Koise and Sakura rivers using a time-integrated SS sampler developed by Phillips et al. (2000). Hourly discharge and rainfall were observed by the Kasumigaura River Office. Daily precipitation data at Tsuchiura weather station (that is, near the outlet of the Sakura River)

were obtained from the JMA (2017). The watershed areas at the sampling points of the Koise and Sakura rivers were 227 and 353 km^2, respectively. The sum of both watershed areas covers 41% of the watershed area of Lake Kasumigaura. The land cover of the experimental catchments is listed in Table 1.

The time-integrated SS sampler was evaluated for its ability to collect a representative particle size of sediment transported in a river system (Smith and Owens, 2014). The maximum efficiency of the sampler for collecting sediment mass was 43% for fine-grained sediment samples and 87% for coarser-grained samples. To measure the in situ particle size distribution in the Sakura River, the LISST-StreamSide (Sequoia Scientific Inc.) was deployed from August −;September 2015. The LISST-StreamSide is practical for use in either laboratory or the field and uses laser diffraction technology to estimate particle size class concentration metrics (Hubbart et al., 2014; Landers and Sturm, 2013). Rainfall of 358 mm occurred during the deployment period, accounting for 30% of the annual precipitation in 2015. Particle size ranges from 1.9 to 387 μm were scanned and classified into 32 categories. Additionally, SS collected by the sampler during three periods (March–June, July–August, and August–September 2015) were classified into five categories by wet-sieving with mesh sizes of 425, 250, 106, and 75 μm.

The wet SS samples were then freeze-dried and homogenized. Radioactive concentrations of ^{134}Cs and ^{137}Cs, loss on ignition, and median particle diameter were analyzed in the same way as the sediment samples. Their radioactive concentrations of were analyzed after freeze-drying. The radioactive decay from Mar 15, 2011, was corrected.

Particulate size affects ^{137}Cs concentrations in soils and sediments, increasing with decreased particulate size (He and Walling, 1996). To normalize the particle size dependency of ^{137}Cs concentrations in SS for the yearly trend analysis, the concentration was divided by P, which was calculated using the following equation:

$$P = (X / X_{cr})^{v} \quad (1)$$

where X and X_{cr} are the specific surface areas of each sample and the criteria sample, respectively. The specific surface area was estimated using the particulate size distribution of the sample and the spherical approximation of particles in each size class. The S_s was determined to be 0.65 $m^2\ g^{-1}$ from the SS collected in the Koise River during September–October 2012. The exponent coefficient v was constant. He and Walling (1996) estimated the coefficient v for various types of soil and sediment samples and reported a relatively constant v value ranging from 0.65 to 0.75. In the present study, we applied a v of 0.65, as reported by Yoshimura et al. (2015b) in the Abukuma River basin in Fukushima Prefecture.

After correction of radioactive decay from Mar 15, 2011, the particle size-specific ^{137}Cs concentrations (C', Bq kg^{-1}) were fitted by an exponential decay model represented by the following equation:

$$C' = C_0\ e^{-k\ t} \quad (2)$$

where C_0 (Bq kg^{-1}) and k (y^{-1}) are empirically determined constants and t is the elapsed time since Mar 15, 2011 (y). The effective half-lives, T_{eff} (y), were calculated by dividing the natural logarithm of 2 by the rate of decline, k.

2.5. Estimation of radiocesium accumulation in Lake Kasumigaura

The total amounts of ^{137}Cs accumulated in the entire lake were estimated in March 2011, December 2012, and August 2016. Radioactivity was corrected for decay on Mar 15, 2011.

The ^{137}Cs inventories were obtained based on sediment cores collected at sites 3, 9, and 24 in June 2007 (Table 2; Fukushima et al., 2010). Assuming that these three sites represented the area partitioned by the double dotted lines in Fig. 1 corresponding to Takahama-iri Bay area of 23 km^2, Tsuchiura-iri Bay area of 49 km^2, and the other area of 99 km^2, the total pre-accident amount of ^{137}Cs could be estimated. This value was regarded as the amount in March 2011, assuming negligible input or output of ^{137}Cs until March 2011.

Tsuji et al. (2019) investigated the ^{137}Cs inventory at a depth of 0–15 cm in December 2012 based on sediment cores collected from 68 points covering the entire lake. From the spatial-interpolated data, the accumulated amount of ^{137}Cs from a depth of 0–15 cm in the whole lake (M_{15}) could be calculated as 3.1×10^{12} Bq. In the present study, the total amount of ^{137}Cs (M_{total}) was estimated by multiplying M_{15} by the correction ratio. This ratio was 1.2 using cores >40 cm collected at sites 3, 9, and 24 in November 2013 by assuming the regional representativeness of these sites. The ratios were nearly unchanged at the center of the lake (site 24) between July 2012 and November 2013.

^{137}Cs inventories were obtained at 25 sites in August 2016 and at the other eight sites in September 2015. For the eight sites, the inventories in 2015 were multiplied by the mean ratio of the inventory in 2016 to 2015 for the three sites. The horizontal distribution was estimated by applying spline interpolation to the inventories at the 33 sites. The total ^{137}Cs amount was calculated from the interpolated data.

2.6. Estimation of loads of particulate radiocesium inflowing to Lake Kasumigaura

The entrainment coefficient for solid wash-off, S_c ($m^2\ kg^{-1}$), defined as the ratio of the ^{137}Cs concentration in SS to the ^{137}Cs deposition in the catchment area (Garcia-Sanchez and Konoplev, 2009), was calculated for the Koise and Sakura rivers. The catchment area-weighted average S_c (S_c') was then determined. The time-series of S_c' was approximated by Eq. (2). The riverine input of particulate ^{137}Cs from the entire catchment to the lake, I_p (Bq), was estimated using the following equation:

$$I_p = \alpha \int D\ S_c'\ L_{SS}\ dt, \quad (3)$$

where α is the correction factor; D is the ^{137}Cs deposition amount in the entire catchment of the lake (Bq m^{-2}); and L_{SS} is the SS load from the whole catchment (kg y^{-1}).

α was used to correct the influence of the bias of the particle size of SS collected by the sampler on the ^{137}Cs concentration. The time-integrated SS sampler may not be suitable for collecting representative samples from large rivers because of uncollected fine particles (Smith and Owens, 2014), and particulate size affects ^{137}Cs concentrations in soils and sediments (He and Walling, 1996). The α was determined in the Sakura River during August–September 2015, the deployment period of the LISST-StreamSide, as shown in the following equation:

$$\alpha = \sum f_i\ C_i / \sum g_i\ C_i \quad (4)$$

where f_i and g_i are the proportion of each particle size range from in situ measurements by the LISST-StreamSide and laboratory measurements by the wet-sieving method, respectively; and C_i is the ^{137}Cs concentration of each sieved sample. The subscript i indicates the particle size class of the four categories (<75, 75–106, 106–250, and >250 μm). Because the particle size distribution of the LISST-StreamSide is volume-based, it was assumed that the specific gravity was constant, despite the particle size.

The L_{SS} was first estimated using the relationship between L_{SS} and water discharge (L-Q equation) in the experimental catchment and the ratio of the entire catchment area to the experimental catchment area. However, the estimated L_{SS} was approximately one-third of the mass sedimentation rate in the lake, as reported by Fukushima et al. (2010). The L-Q equation did not include bed load transportation and may have a large error during flooding, causing an underestimation of the L_{SS}. Therefore, the mass sedimentation rate of the lake was used for calculating the L_{SS}.

2.7. Laboratory experiment predicting solid-liquid partition coefficient of radiocesium

The solid-liquid partitioning coefficient of ^{137}Cs, K_d (L kg^{-1}), was determined by laboratory experiments. A sediment sample with a relatively high ^{137}Cs concentration was used for the experiment (that is, from site 16 in July 2012 at a 6–8-cm depth). Lake water was collected at site 24 in February 2010 prior to the FDNPP accident; therefore, the dissolved ^{137}Cs concentration was assumed to be zero. The sediment (1 g) was mixed with filtered water (10 L) and stirred continuously for 1 d. The dissolved ^{137}Cs concentration was analyzed after filtering through 0.7-μm glass fiber and heat condensation. K_d was estimated as the ratio of the ^{137}Cs concentration in sediment to that in water, assuming equilibrium conditions at the end of the experiment.

3. Results

3.1. Temporal and spatial changes in the radiocesium concentration in Lake Kasumigaura sediments

Similar profiles of sediment properties (water content, specific gravity, apparent density, and loss on ignition) were collected during the sampling years (Fig. S1). The water content decreased with depth in the sediment cores, and loss on ignition showed a similar tendency at several sites. In contrast, apparent density generally increased with depth, and specific gravity ranged from 2.0 to 2.5 and did not exhibit clear tendencies. Compared with the center of the lake, the water content was notably lower at several sites near the shore. In contrast, the median diameters of the sediment particles were relatively large near the shore (e.g., sites 1, 5, and 33; Table S1).

The vertical profiles of ^{137}Cs concentrations in the sediment cores taken at six sites are shown in Fig. 2. The profiles in 2007 (that is, before the FDNPP accident) are also shown at sites 3, 9, and 24 (<33 Bq kg^{-1}; Fukushima et al., 2010).

At the center of the lake (site 24), the ^{137}Cs peak could be found at the sediment surface (0–2 cm) 6 months after the accident (September 2011; 1100 Bq kg^{-1}). However, vertical homogenization was observed over time and an approximately flat profile was found at a depth of 0–20 cm within 5.5 years of the accident (August 2016; approximately 430 Bq kg^{-1}). The ^{137}Cs concentration decreased exponentially with depth in the lower layer. The details of the vertical ^{137}Cs profiles at the center of the lake are described in Fukushima et al. (2018).

Vertical homogenization was also observed at sites 1 and 3, located in the Takahama-iri Bay. Notably, at site 3, flat profiles were found at a

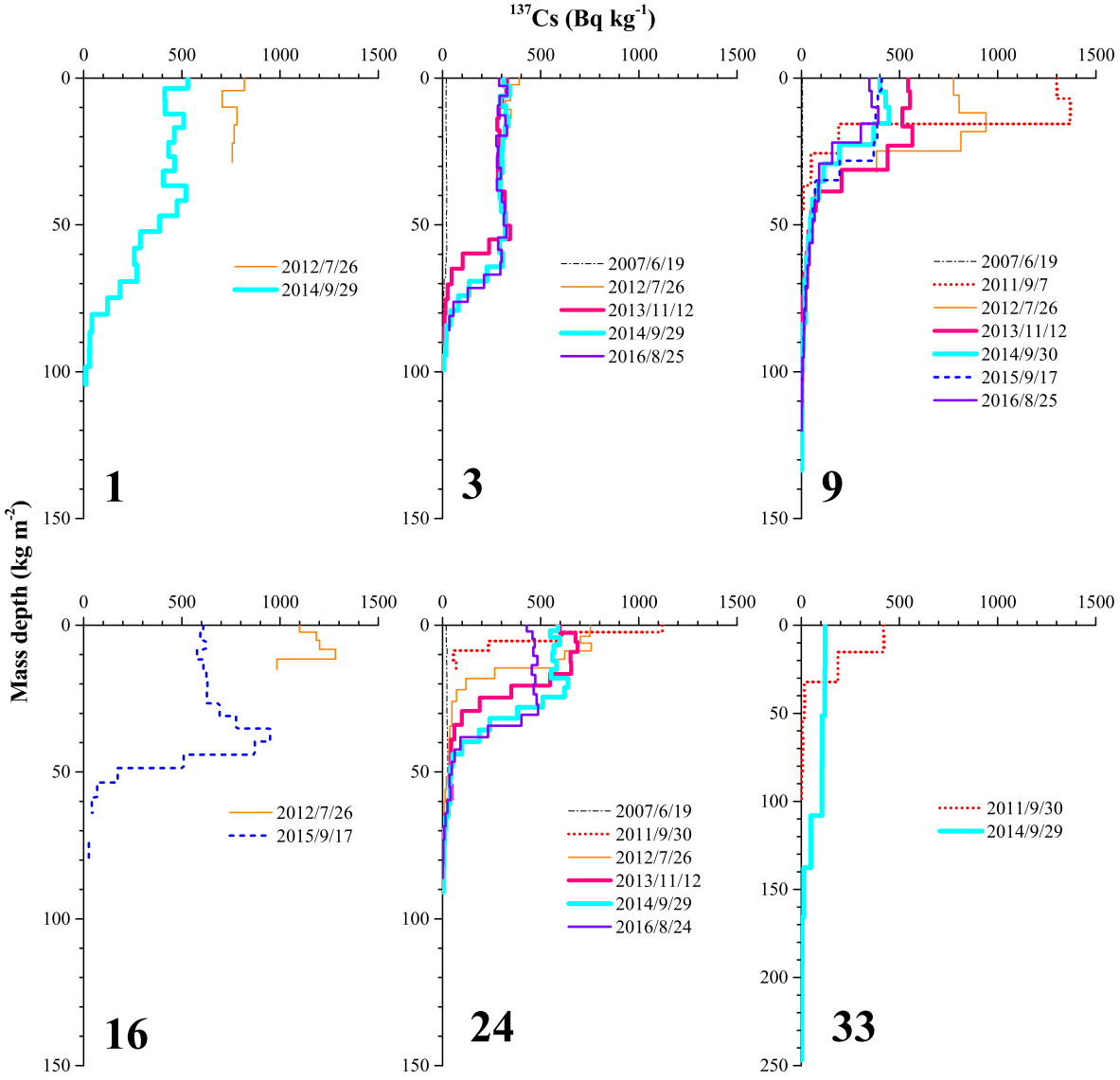

Fig. 2. Vertical profiles of ^{137}Cs in the sediment cores taken at six sites during 2007 and 2011–2016.

depth of 0–30 cm during 2013–2016 (approximately 300 Bq kg^{-1}). However, different features were found at sites 9 and 16, located in Tsuchiura-iri Bay. At site 9, ^{137}Cs did not reach the deep sediment. At site 16 (between sites 9 and 24), the peak ^{137}Cs concentration derived from the FDNPP was observed at a depth of 20–22 cm in 2015. Some of our results are consistent with the results of Tsuji et al. (2019), in which ^{137}Cs profiles at sites 3, 9, and 24 during 2012–2013 were reported.

A decline in the ^{137}Cs concentration at the sediment surface was observed (Fig. 3). Especially at site 9 (in Tsuchiura-iri Bay), the concentration at the sediment surface decreased significantly from 1300 Bq kg^{-1} in 2011 to approximately 400 Bq kg^{-1} in 2014–2016. Compared to other sites, the concentration at site 3 did not change noticeably during 2012–2016; however, no observations were made in 2011. The ^{137}Cs concentration decreased over the 6 years following the accident; however, it remained one or two orders of magnitude higher than that before the accident.

The rate of decline in the ^{137}Cs concentration was estimated using Eq. (2) after radioactive decay correction to 0.05 y^{-1} at site 3 in Takahama-iri Bay ($r^2 = 0.73$). However, Fig. 3 shows that the decrease in the ^{137}Cs concentration could be divided into two periods at sites 9 and 24. For approximately 3 years after the accident, the rates of decline of the ^{137}Cs concentration were 0.40 y^{-1} at site 9 and 0.29 y^{-1} at site 24; after which the rate decreased to 0.13 y^{-1} at site 9 and 0.10 y^{-1} at site 24. Matsuzaki et al. (2021) fitted the two-component models to the concentrations of dissolved ^{137}Cs in Lake Kasumigaura and found a higher rate of decline (k_2 of 0.47) than our study in terms of sediment decline.

3.2. Temporal and spatial changes in the radiocesium inventory in lake sediments

After the FDNPP accident, the ^{137}Cs inventory increased from 5.7 kBq m^{-2} in 2011 to 23 kBq m^{-2} in 2014 at site 24 (that is, around the center of the lake) (Fig. 4a). Increasing trends were also confirmed at sites 3 (Takahama-iri Bay) and 33 (near the outflow). In contrast, a decrease in the ^{137}Cs inventory was observed at site 9 (located in Tsuchiura-iri Bay) during 2012–2014. After November 2014, few changes were observed at most of the observation sites. The ^{137}Cs inventories at sites 3, 9, and 24 in July 2007 (before the accident) were 0.4–1.6 kBq m^{-2}, one or two orders of magnitude lower than those after 2011.

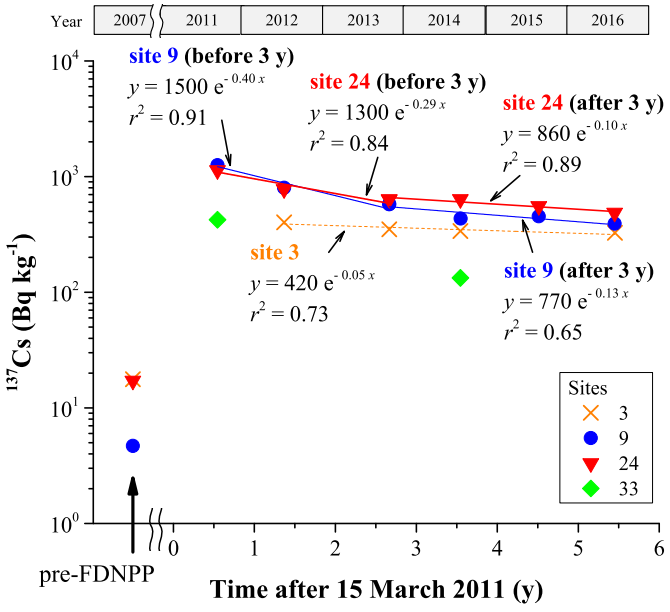

Fig. 3. Changes in the ^{137}Cs concentration at the sediment surface at four sites during 2011–2016. The ^{137}Cs concentration in June 2007 is shown as "pre-FDNPP." Radioactive decay from Mar 15, 2011, was corrected. FDNPP - Fukushima Dai-ichi Nuclear Power Plant.

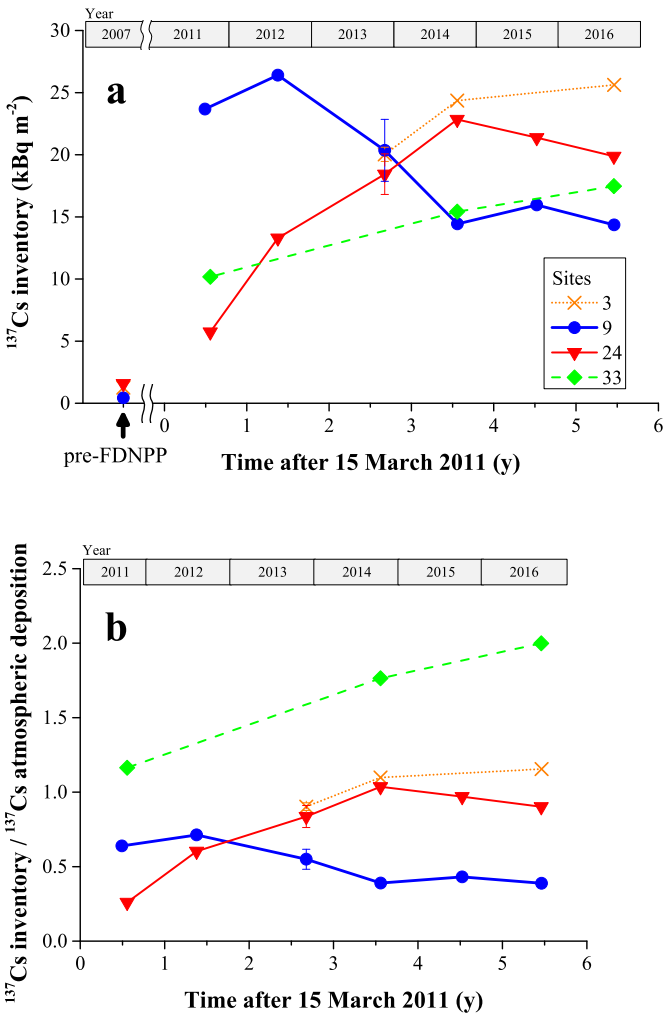

Fig. 4. (**a**) Changes in the ^{137}Cs inventory in the sediment cores and (**b**) the ratio of the ^{137}Cs inventory to the initial ^{137}Cs deposition at four sites during 2011–2016. Radioactive decay from Mar 15, 2011, was corrected. In (a), the ^{137}Cs inventory in the sediment core taken in June 2007 is shown as "pre-FDNPP." FDNPP - Fukushima Dai-ichi Nuclear Power Plant.

The ratios of the ^{137}Cs inventory in sediments to the initial ^{137}Cs deposition are shown in Fig. 4b. At the lake center (site 24), the ratio was 0.26 in September 2011; however, the ratio was approximately 1 after 2014. At site 3, the ratios were similar to those at site 24. At site 33, the ratios were >1 and increased during the observation period. In contrast, the ratios were <1 and decreased at site 9 (located in Tsuchiura-iri Bay). Yoshimura et al. (2014) showed ratios in the range 0.19–1.33 in four irrigation ponds 4–5 months after the FDNPP accident. In the present study, the ratios determined in 2011 were within the values reported by Yoshimura et al. (2014).

The ^{137}Cs inventory in lake sediments was spatially interpolated, mainly based on observation data from August 2016 (Fig. 5a). The ^{137}Cs accumulated intensively around the outlet of the inflowing rivers, such as the Koise, Ono, and Hanamuro rivers. However, the inventory was not high around the Sakura River outlet, which has the largest watershed among all inflows. As a reference, Tsuji et al. (2019) investigated the spatial distribution of ^{137}Cs in the lake during 2012–2014, indicating that the ^{137}Cs inventory was likely to increase near urban river mouths. Compared to 2014 (Tsuji et al., 2019), ^{137}Cs accumulation progressed at the mouth of several inflows in 2016. Additionally, the inventory

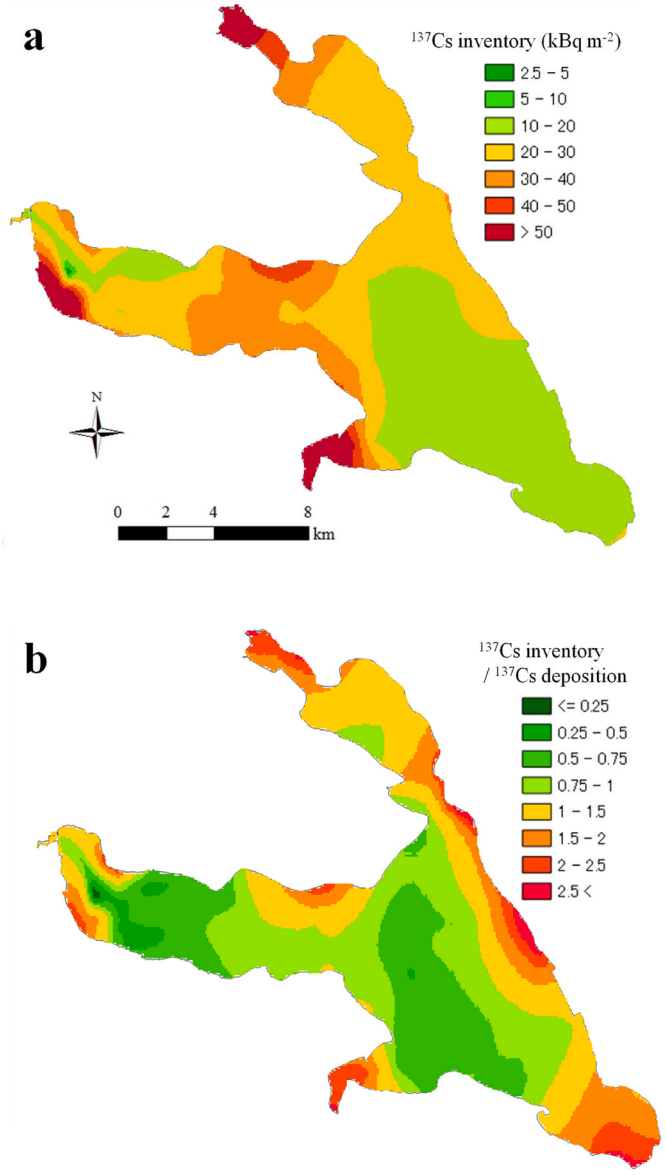

Fig. 5. (**a**) Spatial distribution of the ^{137}Cs inventory and (**b**) the ratio of the ^{137}Cs inventory to the initial ^{137}Cs deposition covering the entire lake in August 2016. Sediment cores taken at 25 sites in 2016 and eight sites in 2015 were used for inventory calculation. Radioactive decay from Mar 15, 2011, was corrected.

increased around the bay mouth of Tsuchiura-iri Bay since 2014. The inventory was relatively low in the southeastern part of the lake, far from inflowing rivers.

The spatial distribution of the ratios of the ^{137}Cs inventory in the sediments (Fig. 5a) to the initial ^{137}Cs deposition (Fig. 1) is shown in Fig. 5b. The ratio was <1 from Tsuchiura-iri Bay (high deposition area; Fig. 1) to the center of the lake. The ratio was >1 and relatively higher around the outlet of the main inflows, except near the outlet of the Sakura River flowing to Tsuchiura-iri Bay. The ratio was also higher around the outflow (low-deposition area; Fig. 1).

3.3. Changes in the particulate radiocesium concentration in two main inflows

Riverine SS (0.3–220 g) was collected by the time-integrated SS sampler for each sampling period. Detailed analytical results are presented in Table S2 The concentration of ^{137}Cs weighted average by SS weight was 430 Bq kg^{-1} in the Koise River, which was higher than that in the Sakura River (84 Bq kg^{-1}) during September 2012–December 2015. It was consistent with the mean ^{137}Cs deposition in the catchments of the Koise (21.4 kBq m^{-2}) and Sakura rivers (7.8 kBq m^{-2}) (Table 1). ^{137}Cs concentrations of SS in the two rivers in 2009 were 4–6 Bq kg^{-1}, one or two orders of magnitude lower than those during 2012–2015. The ^{137}Cs concentration was considerably high in the sample collected from February to March 2013 in the Sakura River; however, the minimum SS weight was also recorded. This observation had a small effect on the amount of loading in the lake but a large effect on the trend analysis, and could be an error. Therefore, we excluded this finding from the trend analysis.

By fitting the exponential decay model expressed in Eq. (2) after the particle size correction, k and T_{eff} were determined to be 0.39 y^{-1} and 1.8 y in the Koise River, and 0.30 y^{-1} and 2.3 y in the Sakura River, respectively (Fig. 6a). Significantly decreasing trends in ^{137}Cs concentrations in SS were detected in both rivers ($p < 0.001$). Saxén (2007) reported a decreasing trend of ^{137}Cs concentrations in Finnish lake water samples during 1986–2001, which could be divided into three stages: Rapid decrease after deposition ($k_1 = 5$ y^{-1}), second highest decrease ($k_2 = 0.7$ y^{-1}), and very slight decrease ($k_3 = 0.1$ y^{-1}); the rate k in the present study was between k_2 and k_3. In Fukushima Prefecture, Iwagami et al. (2017) reported the k value for particulate ^{137}Cs to be 0.75 y^{-1} in a

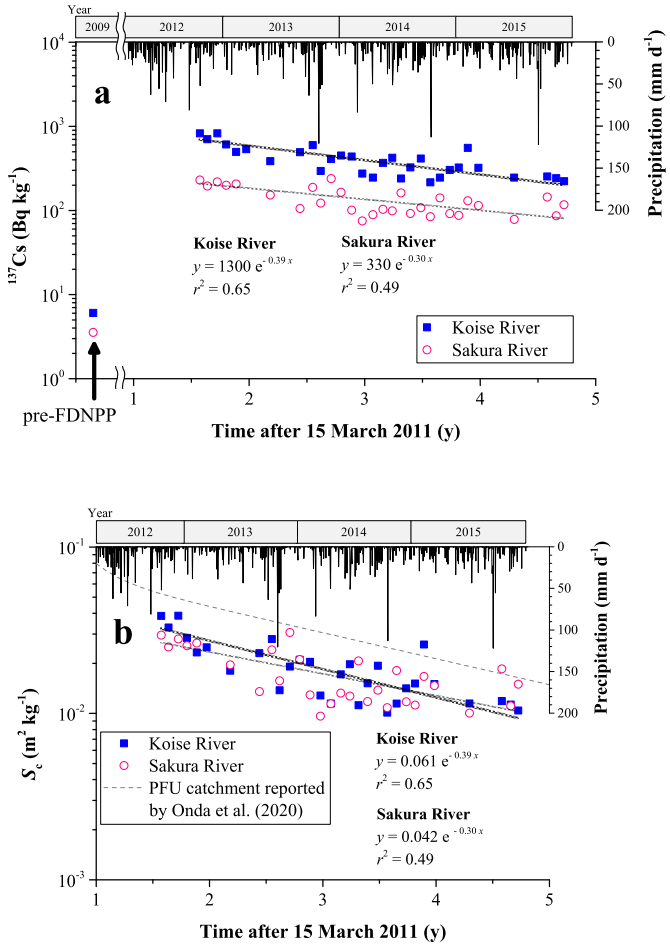

Fig. 6. (**a**) Changes in the particle size-specific ^{137}Cs concentration of suspended solids (SS) and (**b**) the entrainment coefficient of ^{137}Cs, S_c, in the Koise and Sakura rivers from September 2012–;December 2015. Radioactive decay from Mar 15, 2011, was corrected. Daily precipitation at Tsuchiura weather station is also shown. In (a), the ^{137}Cs concentration of SS collected in November 2009 is shown as "pre-FDNPP." FDNPP - Fukushima Dai-ichi Nuclear Power Plant.

headwater catchment from August 2012−;September 2013. The values observed in the present study were approximately half of the previously reported values.

The ^{137}Cs concentrations in SS were positively related to a loss on ignition of SS in the Koise ($p < 0.05$) and Sakura rivers ($p < 0.01$), although a temporal trend of loss on ignition was not detected (Fig. S2).

SS samples collected by the time-integrated sampler were classified into five particle size categories, and the ^{137}Cs concentrations are shown in Table 3. The SS sample with a particle size <75 μm had the largest weight percentage among the five particle size categories in both rivers during each deployment period. The ^{137}Cs concentrations in the fine-grained sample were higher than those in the coarse-grained sample, especially during August–September 2015 when the maximum SS weight was recorded among the three deployment periods due to rainfall events.

The weight percentage of SS collected by the time-integrated sampler was compared with the volume percentage of SS measured by the LISST-StreamSide with classification into four particle size categories in the Sakura River from August−;September 2015 (Table 4). The highest concentration was observed in the <75 μm-class. Assuming that the specific gravity of SS was constant for several particle sizes, the percentage of this fraction was 44% by the sampler, which was lower than that by the LISST-StreamSide (60%). The correction factor α was calculated to be 1.24 using Eq. (4).

3.4. Solid–liquid partition coefficient of radiocesium in Lake Kasumigaura

The laboratory experiment showed that the K_d of ^{137}Cs was 7.0 × 10^4 L kg^{-1}. Previous studies have reported K_d values in the range of 2–60 × 10^4 L kg^{-1} in various lakes (Ilus and Saxén, 2005; Konoplev et al., 2002; Putyrskaya et al., 2009), 0.6–500 × 10^4 L kg^{-1} around Fukushima Prefecture (Nagao et al., 2013; Sakaguchi et al., 2015; Ueda et al., 2013), and 1.6–3.4 × 10^4 L kg^{-1} in Lake Kasumigaura (Tsuji et al., 2019). The present K_d value was within or close to these value ranges.

3.5. Accumulation amounts and inflowing loads of particulate ^{137}Cs in Lake Kasumigaura

Table 5 shows the accumulation amounts and inflowing loads of particulate ^{137}Cs in the lake. The total amount of ^{137}Cs was estimated to be 0.20 × 10^{12} Bq in 2007. This value could be regarded as the amount that was present just before the FDNPP accident because of the negligible input of ^{137}Cs confirmed by SS samples in 2009. The total amount of ^{137}Cs in December 2012 was estimated to be 3.72 × 10^{12} Bq. The spatial distribution of the ^{137}Cs inventory in August 2016 (Fig. 5a) revealed that the total amount of ^{137}Cs was estimated to be 4.18 × 10^{12} Bq. Therefore, the amount increased by 3.52 × 10^{12} and 0.46 × 10^{12} Bq during March 2011–December 2012 (first 1.8 y) and December 2012–August 2016 (second 3.7 y), respectively. The mean accumulation rates were 2.0 × 10^{12} and 0.13 × 10^{12} Bq y^{-1} during each period, respectively.

The riverine input of particulate ^{137}Cs from the entire watershed area from December 2012−;August 2016 was estimated to be 0.47 × 10^{12} Bq with no correction and 0.58 × 10^{12} Bq with sampler bias-correction (that is, by applying the correction factor α), which was similar to the increase in sediments of 0.46 × 10^{12} Bq during the same period (Table 5). The estimated riverine input during this period accounted for 1%–2% of the ^{137}Cs deposition in the catchment and 11%–14% of the amount of ^{137}Cs accumulated in the lake in 2016.

Table 3
Particle size distributions and ^{137}Cs concentrations of sieved suspended solid (SS) samples collected in the Koise and Sakura rivers during the three deployment periods (March–June, July–August, and August–September 2015).

Sampler setting date	Particle size (μm)	Koise River			Sakura River		
		SS weight		^{137}Cs (Bq kg^{-1})	SS weight		^{137}Cs (Bq kg^{-1})
		(g)	(%)		(g)	(%)	
2015/3/23	<75	15.2	84	290	8.5	53	71
	75–106	0.84	05	330	1.7	11	65
	106–250	1.7	10	280	3.8	23	30
	250–425	0.17	01	320	1.6	10	34
	425<	0.05	00	–	0.52	03	39
2015/7/9	<75	12.9	76	310	21.8	63	72
	75–106	1.8	11	300	3.7	11	48
	106–250	2.1	12	320	7.7	22	66
	250–425	0.09	01	170	1.1	03	58
	425<	0.07	00	–	0.46	01	61
2015/8/13	<75	23.1	74	260	66.6	44	88
	75–106	2.8	09	190	9.4	06	32
	106–250	4.4	14	140	43.4	28	15
	250–425	0.70	02	130	19.1	13	15
	425<	0.29	01	059	14.0	09	8.9

Table 4
Comparisons of weight percentage of suspended solids (SS) collected by the time-integrated sampler with volume percentage of SS measured by the LISST-StreamSide in the Sakura River during August–September 2015.

Particle size (μm)	Time-integrated SS sampler			LISST-StreamSide
	SS weight		^{137}Cs (Bq kg^{-1})	Percentage of SS volume (%)
	(g)	(%)		
<75	66.6	44	88	60
75–106	9.4	06	32	04
106–250	43.4	28	15	11
>250	33.1	22	13	25

Table 5
Accumulation amounts and inflowing loads of particulate ^{137}Cs in Lake Kasumigaura.

Period	(10^{12} Bq)	(10^{12} Bq y^{-1})
Accumulation in the entire lake (based on sediment analysis)		
Mar. 2011–Dec. 2012	3.5	2.0
Dec. 2012–Aug. 2016	0.46	0.13
Input from rivers as particulate form (based on SS analysis)		
Dec. 2012–Aug. 2016	0.47–0.58	0.13–0.16

4. Discussion

4.1. Characteristics of the entrainment coefficient of particulate radiocesium in the Koise and Sakura rivers

The ^{137}Cs concentrations of SS from the Koise River were greater than those from the Sakura River (Fig. 6), which corresponded to the deposition in the catchment. In the Lake Kasumigaura basin, Tabayashi and Yamamuro (2013) found a correlation between radiocesium concentrations in the riverbed sediment and the amount of radiocesium deposited in the upstream catchment, which was consistent with our results.

The S_c values ranged from 0.01 to 0.04 m^2 kg^{-1} in the Koise River during September 2012–December 2015, which were slightly higher than the range in the Sakura River (0.009–0.03 m^2 kg^{-1}); however, the S_c values in the Sakura River had uncertainty because the deposition in some watersheds was below the detection limit (<5 kBq m^{-2}; Fig. 1). These S_c values were within the range of 0.004–0.08 m^2 kg^{-1} reported from farmland, grassland, and forest in Fukushima Prefecture by Yoshimura et al. (2015a), indicating that forests had higher S_c values than other land uses (such as grassland and farmland) during July 2011–November 2012. For farmlands, the effect of cultivation on lowering the S_c was suggested.

Onda et al. (2020) constructed the S_c trendline of the land use related to human activities (paddy fields, farmland, and urban areas: PFU), which is almost consistent with the slope of the S_c in the present study (Fig. 6b). However, there was an approximately two-fold difference in the S_c value between them, which may have been because of the characteristics of the target area (Lake Kasumigaura basin), such as smaller initial deposition or large initial migration of deposition due to larger anthropogenic influence.

The S_c was initially higher in the Koise River but decreased exponentially in the Koise River than in the Sakura River until 2015. Therefore, the difference in ^{137}Cs concentrations will probably be smaller in the future. The considerable S_c decrease in the heavily forested Koise River catchment was consistent with that reported by Onda et al. (2020).

4.2. Comparisons of the ^{137}Cs concentration decline rate between river SS and Lake Kasumigaura sediments

Declines in the ^{137}Cs concentration were observed in both river SS and lake bottom sediments (Figs. 3 and 6). The rates of decline k in lake sediments at a depth of 0–2 cm (0.05–0.40 y^{-1} from three sites) were lower than those in river SS (0.30–0.39 y^{-1} from two rivers), which may have been caused by post-depositional processes in the lake.

A difference in the decline rates was observed between lake sediments at site 3 in Takahama-iri Bay (0.05 y^{-1}) and SS in the Koise River flowing to the bay (0.39 y^{-1}). At site 3, the ^{137}Cs concentration in the surface sediments was nearly constant at 300–400 Bq kg^{-1} during the observation period (Fig. 3). In the Koise River (Fig. 6), the ^{137}Cs concentration of SS was greater than 600 Bq kg^{-1} in 2011–2013 and decreased to a concentration of less than 300 Bq kg^{-1} by 2015. Thus, the ^{137}Cs concentration in lake sediments was more stable than that in river SS, suggesting the effect of vertical disturbance of lake sediments on the ^{137}Cs concentration.

Rapid migration of ^{137}Cs to a certain depth was observed for a few years since 2011, especially at site 3 (Fig. 2). This could not be explained by the mass sedimentation rate reported by Fukushima et al. (2010) of approximately 1 kg m^{-2} y^{-1}. The ^{137}Cs intrusion depth suggests post-depositional processes, such as disturbance by waves, bioturbation, and remobilization due to water quality changes. Erlinger et al. (2008) observed the vertical disturbance of sediment ^{137}Cs at shallow water depths, which is consistent with the Lake Kasumigaura characteristics (mean water depth of 4 m). Fukushima et al. (2018) described the penetration observed at the center of Lake Kasumigaura using a one-dimensional differential sediment model. They concluded that wind-induced stress and sediment porosity were the key parameters determining the observed profiles in the lake. Tsuji et al. (2019) reported that rapid penetration was observed not only at the center but also in Takahama-iri Bay, which was consistent with the results of the present study.

The importance of post-depositional processes was indicated with respect to the long-term fate of ^{137}Cs in a drinking water reservoir (Kaminski et al., 1997). In Lake Kasumigaura, the vertical mixing caused a rapid decline of ^{137}Cs concentration in the sediment surface after the accident. However, the ^{137}Cs concentration may decline more slowly than in other lakes from a long-term perspective because the vertical mixing masks the effect of new deposition with low ^{137}Cs concentration in the future. Therefore, it is expected that the conditions under which ^{137}Cs is dissolved/resuspended will be prolonged, whereas the influence of ^{137}Cs outflow because of dissolution/resuspension on the mass balance of ^{137}Cs in the lake may be small.

However, small differences in the decline rates of the ^{137}Cs concentration were observed between lake sediments at site 9 in Tsuchiura-iri Bay (0.13–0.40 y^{-1}) and SS in the Sakura River flowing to the bay (0.30 y^{-1}). At site 9, the sediment was less vertically mixed than at site 3 (Fig. 2), which may have caused the different decline rates between the two bays. The ^{137}Cs concentration in surface sediments at site 9 (>400 Bq kg^{-1} until 2015; Fig. 3) was higher than that in SS collected from the Sakura River (mostly <200 Bq kg^{-1}; Fig. 6) during the observation period, suggesting the contribution of high depositions of ^{137}Cs around the southwest of Lake Kasumigaura basin, including Tsuchiura-iri Bay (Fig. 1).

4.3. Horizontal migration of radiocesium mainly carried by fine particles in Lake Kasumigaura

The ^{137}Cs inventories at the center of the lake (site 24) appeared to increase monotonically until 2014 (Fig. 4a), indicating ^{137}Cs transport from watersheds/airsheds or upstream sediments. Fig. 5 demonstrates that ^{137}Cs accumulated around the outlet of the inflowing rivers, as also reported in previous studies (Blakar et al., 1992; Tsuji et al., 2019).

We confirmed the decreasing trend of the ^{137}Cs inventory at site 9 in Tsuchiura-iri Bay (Fig. 4a). The ratio of the ^{137}Cs inventory to deposition was <1 in this area in 2016 (Fig. 5b). These results suggest resuspension and flow of sediments, especially fine particles. The median diameter (d_{50}) of the sediment surface at site 9 (7.6 μm) was larger than that at sites 3 (4.8 μm) and 24 (4.1 μm), which may have resulted from fine particles flushing out from site 9 (Table S2). Therefore, this area of Tsuchiura-iri Bay may be a source of ^{137}Cs in the downstream area, contributing to the observed increasing trend of the ^{137}Cs inventory around the lake center (Fig. 4a). This characteristic observation was only made in Tsuchiura-iri Bay and not in Takahama-iri Bay, which may have been influenced by differences in bay geography. It was reported that the exchange flow discharge between Tsuchiura-iri Bay and the center area of the lake was much larger than that between Takahama-iri Bay and the central area due to bay geography (Arai and Fukushima, 2014). The significant transport of particulate ^{137}Cs may have been attributed to the lake's shallowness, which enhances the resuspension of bottom sediment during strong winds (Asai and Tsubogo, 2006). For example, Koibuchi et al. (2015) reported notable transport of particulate ^{137}Cs around Lake Teganuma (average depth of 0.9 m) from October 2011−; March 2014 based on field observations.

Tsuji et al. (2019) reported changes in the ^{137}Cs inventory in the lake until 2014, such as a decrease in Tsuchiura-iri Bay and an increase around the center of the lake, which is consistent with our results. Since 2014, however, a significant increase/decrease in the ^{137}Cs inventory was not observed at several points in the lake (Fig. 4a). This indicates an effect of the surface ^{137}Cs concentrations approaching a constant level across several sites in recent years (Fig. 3). The coefficient of variance of the ^{137}Cs inventory among sites 9, 24, and 33 was 58% in September 2011, and decreased to 23% in 2014 and 13% in 2016. This trend suggests a spatial homogenization of the inventory by migration of ^{137}Cs after accumulation, such as resuspension and flow of fine particles. This suggestion is consistent with Naya et al. (2004), who measured the horizontal distribution of grain size in Lake Kitaura, located next to Lake Kasumigaura (Nishiura). The authors considered that the fine sediments were eroded by wave action near the shore of the lake, then transported by suspension and settled in the central region of the lake.

Besides the above considerations, the amount of ^{137}Cs loading from the watershed is expected to decrease. Therefore, the difference in the ^{137}Cs inventory between the northern parts (near the inlets) and southern parts (near the outlet) of the lake will decrease.

4.4. Budgetary estimation of radiocesium in Lake Kasumigaura during December 2012–August 2016

The change in ^{137}Cs accumulated in the lake, ΔM (Bq), can be expressed by the following formula:

$$\Delta M = \mathrm{A}\ Tr + I_p + I_d - O \quad (5)$$

where A is the atmospheric deposition of ^{137}Cs on the lake surface (Bq); *Tr* is the trap efficiency; I_d is the riverine input in dissolved form (Bq); and *O* is the output through outflow (Bq). The experimental catchment for estimation of the riverine input had a higher percentage of forest area and a lower percentage of paddy, crop, and urban areas than the entire catchment (Table 1), which may have caused an error in the estimation. However, our observations revealed that the I_p of 0.47–0.58 × 10^{12} Bq was similar to the *ΔM* of 0.46 × 10^{12} Bq in the lake during December 2012–August 2016, suggesting that the error was small. Therefore, the net influence of other paths (*A Tr* + I_d – *O*) on the accumulation (*ΔM*) may have been sufficiently small.

The contribution of atmospheric input of ^{137}Cs was relatively small after December 2012 from monthly observation data of atmospheric ^{137}Cs deposition at the MRI, approximately 10 km west of the lake (Igarashi et al., 2015). Using these data, the *A* was estimated to be 4.4 × 10^{10} Bq.

Some of the ^{137}Cs initially deposited on the lake surface may have flowed out of the lake without reaching the bottom due to water surface tension. The ratios of the sediment ^{137}Cs inventory in September 2011 to the initial ^{137}Cs atmospheric deposition were 0.64 at site 9 and 0.26 at site 24 (Fig. 4b; note that site 33 was excluded because of its low spatial representativeness). Therefore, the *Tr* was estimated to be 0.45, as the mean value among the two sites. Thus, the *A Tr* was estimated to be 2.0 × 10^{10} Bq, accounting for <5% of the *ΔM* during this period.

The I_d was estimated to be approximately 2.3 × 10^{10} Bq during December 2012–August 2016 using the K_d value measured by our laboratory experiment, the SS concentration in the rivers, and the total water discharge (Arai and Fukushima, 2012). Additionally, assuming that the K_d in the Abukuma River systems (9–200 × 10^4 L kg^{-1}) reported by Yoshimura et al. (2015b) could be applied to the Lake Kasumigaura basin, I_d was estimated to be 0.082–1.8 × 10^{10} Bq. The estimated values of I_d accounted for <4% of *ΔM* during this period.

The estimated values of *A Tr* + I_d accounted for <9% of *ΔM*. Considering the upper limit of the counting error in the measurement of radiocesium to be 10%, the contribution of atmospheric deposition and riverine input in a dissolved form to accumulation in the lake might be sufficiently small. The *O* may also have been low. The riverine output of ^{137}Cs in its dissolved form was estimated to be approximately 3 × 10^{10} Bq from December 2012–;August 2016, using the K_d value measured in our laboratory experiment, the ^{137}Cs concentration in the sediment surface at the center of the lake, and the water discharge. This estimation assumed K_d to be constant after December 2012, continuous equilibrium conditions, and vertical homogeneity of dissolved ^{137}Cs due to the shallowness of the lake. To obtain a detailed outflow of ^{137}Cs, it will be necessary to measure the seasonal changes in K_d in response to bottom dissolved oxygen (Matsuzaki et al., 2021) and the ^{137}Cs concentrations in aquatic organisms such as phytoplankton and zooplankton, mostly in easy-elution forms (Mori et al., 2017).

The total amount of ^{137}Cs in the lake might not increase significantly in the future in the absence of extreme flood events. The I_p was estimated to be 1.0–1.3 × 10^{11} Bq in 2015, accounting for <3% of the amount of ^{137}Cs accumulated in the lake in August 2016. The ^{137}Cs concentration in riverine SS may continue to decrease, especially in the Koise River where there was a high decline rate (Fig. 6a). Therefore, amount of ^{137}Cs loading through inflows is predicted to decrease.

4.5. Estimation of atmospheric deposition and riverine inputs of radiocesium to Lake Kasumigaura since March 2011

Our observations revealed that the riverine inputs of ^{137}Cs in particulate form could explain the ^{137}Cs accumulated in the lake during December 2012–August 2016 (Table 5). However, the riverine input, I_p, could not be calculated from March 2011–;December 2012 because of the lack of observations of 137Cs in river SS before 2012. Therefore, we attempted a simple estimation of I_p since March 2011 using two methods: (1) estimation from *ΔM* – *A Tr* and (2) extrapolation of the single exponential fitting curve of the ^{137}Cs concentration.

In the first method, I_p was estimated as *ΔM* – *A Tr* during March 2011–August 2016 by assuming that I_d was comparable to *O* from Eq. 6. The *ΔM* was calculated to be 4.0 × 10^{12} Bq (Table 5). Total atmospheric deposition was estimated as the sum of the initial deposition in March 2011 (Fig. 1) and the deposition that occurred since April 2011 (Igarashi et al., 2015). Finally, *A Tr* and I_p accounted for 54% (2.1 × 10^{12} Bq) and 46% (1.9 × 10^{12} Bq) of *ΔM* during the period, respectively, suggesting comparable rates of these inputs. The I_p accounted for only 6% of the deposition in the watershed area.

In the second method, the single exponential fitting curve in Eq. (2) without particle size correction was extrapolated to estimate the ^{137}Cs concentration of river SS since March 2011. The I_p was estimated to be 1.1 × 10^{12} Bq, accounting for 28% of the ^{137}Cs accumulation from March 2011–;August 2016. However, this estimated result requires a high trapping efficiency for atmospheric deposition. The single exponential model may not be appropriate for describing initial rapid runoff after an accident. Onda et al. (2020) showed a rapid decline in the ^{137}Cs concentration during the first year after the accident. The rate of decline of S_c in the present study was similar to that of this model during the observation period (Fig. 6b).

5. Conclusions

Temporal/spatial changes in the inflow and accumulation of ^{137}Cs were observed in Lake Kasumigaura during 2011–2016. The ^{137}Cs concentration in lake sediments decreased exponentially, and the rate of decline in lake sediments was lower than that in river SS, suggesting the effect of post-depositional processes on the ^{137}Cs concentration. The ^{137}Cs inventory in sediments increased at the center of Lake Kasumigaura during 2011–2014, but decreased in Tsuchiura-iri Bay during 2012–2014, suggesting transfer of accumulated ^{137}Cs (such as resuspension of fine particles due to lake shallowness). In Tsuchiura-iri Bay, the decrease in the ^{137}Cs concentration in the surface sediments was high until 3 years after the FDNPP accident, controlled by initial atmospheric fallout and its downstream migration; however, after the third year, the decrease slowed down because of the ^{137}Cs supply from the river.

The amount of accumulated ^{137}Cs in the entire lake was similar to the riverine input of particulate ^{137}Cs during December 2012–August 2016, suggesting that the lake has a high performance in trapping particulate matter from the basin. The amount of ^{137}Cs accumulated in the lake, as observed in 2016, may have originated from atmospheric deposition and riverine inputs at comparable rates. The ^{137}Cs budgets were roughly estimated; however, to obtain detailed budgets, it will be necessary to measure dissolved ^{137}Cs concentrations and the concentrations in aquatic organisms (including phytoplankton and zooplankton). In Lake Kasumigaura, the ^{137}Cs concentration in the inflows continued to decrease, and post-depositional migration of ^{137}Cs was observed. Therefore, distribution of ^{137}Cs inventory in the lake is expected to show fewer regional differences than at present. These findings provide useful insights for future prediction and management of radiocesium contamination and the effects of riverine inputs in general shallow lakes.

Credit Author Statement

Hiroyuki Arai: Conceptualization, Investigation, Data curation, Formal analysis, Writing – original draft, Takehiko Fukushima: Conceptualization, Writing – review & editing, Supervision, Funding acquisition, Yuichi Onda: Writing – review & editing, Supervision, Funding acquisition, Project administration.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

We are grateful to the Kasumigaura River Office for providing the monitoring data on discharge in the rivers. This work was supported by Grants-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT), Japan (24110006 and 26281039), and ERAN Y-21-01.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2022.115905.

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