**Vertical distributions of Pu and radiocesium isotopes in sediments from
Lake Inba after the Fukushima Daiichi Nuclear Power Plant accident:
source identification and accumulation**

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Abstract

Radiocesium isotopes were released into the environment in the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, which resulted from the massive Tohoku earthquake in March 2011. Lake Inba is located in Chiba Prefecture about 200 km from the FDNPP site and the lake was contaminated by radioactivity from the accident. The present work was carried out in order to investigate the vertical distributions and accumulations of radiocesium and Pu in sediment cores collected during 2011-2015, and to study the migration behaviors of radiocesium and Pu isotopes in the sediments. The observed $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios in the core samples were approximately 1 (decay-corrected to 15 March, 2011), suggesting that sediment samples were contaminated by the FDNPP-released radiocesium. Compared with radiocesium inventories in soil in the area surrounding the Lake Inba, the calculated higher radiocesium inventories of the collected sediment cores indicated that most of the radiocesium was subsequently accumulated in the lake, and that the radiocesium input by atmospheric deposition and input from surrounding soil were greater than the output of the radiocesium. The $^{239+240}\text{Pu}$ activities in the sediments were all within the background level seen before the accident. The atom ratios of $^{240}\text{Pu}/^{239}\text{Pu}$ ranged from 0.175 to 0.210, suggesting that Pu originated from global stratospheric fallout rather than from the FDNPP accident.

1. Introduction

A massive earthquake and consequent giant tsunamis occurred on 11 March 2011 along the northeast coast of Honshu Island, Japan, resulting in serious damage to the Fukushima Daiichi Nuclear Power Plant (FDNPP). Consequently, the FDNPP accident caused serious radioactive contamination in the environment through the atmospheric fallout and leakage or direct discharge of highly contaminated liquid wastes (Buesseler et al., 2011; Hirose, 2012; Yoshida and Kanda, 2012; Zheng et al., 2012a, 2012b). The total amount of ^{137}Cs released to the atmosphere from the FDNPP was estimated to be in the range of 7-20 PBq (Chino et al., 2011; Morino et al., 2011; IAEA, 2015). Intensive studies on ^{134}Cs and ^{137}Cs have been conducted for various environmental samples to investigate the concentration, distribution and transport of these released radiocesium isotopes (Yamamoto, 2012; Miyake et al., 2012; Aoyama et al., 2013; Ishii et al., 2013; Bu et al., 2014a; Tagami and Uchida, 2015).

Compared with the studies regarding radiocesium in environmental samples after the FDNPP accident, studies focusing on the possible released Pu isotopes are limited. The possible release of ^{239}Pu ($T_{1/2} = 24110$ y) and ^{240}Pu ($T_{1/2} = 6561$ y) with a large risk for internal radiation exposure due to their very long radioactive half-lives has become a great concern for public safety as well. Depending on emission sources, Pu isotopic ratios, especially the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio, vary significantly in the environment (Table 1), and the

ratios can be applied as a powerful fingerprint for Pu source identification. Following the FDNPP accident, the released Pu isotopes with high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios were detected in samples of litter, soil (0.303-0.330) (Zheng et al., 2012b), black substances (0.286-0.365) (Yamamota et al., 2014) and aerosols (0.32 ± 0.10) collected around Fukushima Prefecture (Shinonaga et al., 2014). In addition, the total amount of $^{239+240}\text{Pu}$ derived from the FDNPP site was estimated to be around $1.0\text{-}6.4 \times 10^9$ Bq (METI, 2011; Yamamoto et al., 2012; Zheng et al., 2012). However, studies on Pu isotopes, particularly, for lake system after the FDNPP accident in Japan are rather few, thus attention should be paid to the contamination situation by Pu isotopes in lake system sediments.

Regarding the contamination by radiocesium in a lake environment, Yoshimura et al. (2014) assessed the concentrations of ^{137}Cs (6.4-34.9 kBq/kg) in surface sediments from four irrigation ponds within 40-50 km from the northwestern part of the FDNPP site, and they found that fine sediments containing high concentrations of ^{137}Cs appeared to be removed from the system by hydraulic flushing. In contrast, Yamada et al. (2015) reported that the radiocesium contamination from the accident was accumulated in the Ogaki Dam Reservoir of eastern Fukushima Prefecture. In fact, the transport of radiocesium in lacustrine sediments is controlled by multiple factors. Ochiai et al (2013) reported that precipitation and geomorphology of the catchment influenced Fukushima-originated radiocesium discharge in the Takidani-ike and Dojo-ike lakes, in western Japan. For Lake Inba, although the lake is

situated in an urban area and is about 200 km away from the FDNPP site, the radioactive contamination was relatively serious in this region (Mikami et al., 2015; Saito et al., 2015). Up to now, only ^{137}Cs activities were determined in the lake surface sediments of around 800 Bq/kg (decay-corrected to 15 March 2011) (Fukushima and Arai, 2014), moreover, the activities of ^{137}Cs in surface sediments exhibited decrease trends during 2011-2014 (MOE, 2011-2014). However, it is unknown whether the drops in radiocesium activities are due to removal from the lake or to downward migration or sedimentation of new soil particles bearing a low radiocesium activity. Therefore, it is extremely necessary to investigate the inventory and vertical distribution of the radiocesium in Lake Inba.

Four sediment cores were collected from Lake Inba during 2011-2015 and they were used in this work to investigate the vertical distribution of radiocesium and to understand its subsequent behavior. The $^{239+240}\text{Pu}$ activity and atom ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ were also determined in the sediment cores to identify Pu sources.

2. Materials and method

2.1. Sediment cores sampling

Lake Inba in Chiba Prefecture with surface area of 11.55 km² (Fig. 1) and watershed area 541 km², is one of the most eutrophic lakes in Japan and it is about 200 km away from the FDNPP site. The average depth of the lake is around 1.7 m with a maximum depth of 2.5 m,

and storage capacity is around $19.7 \times 10^6 \text{ m}^3$. The residence time of the water is about 22 days (Amano and Nakanishi, 2008). It serves as a prefectural nature park that provides leisure fishing activities and plays a major role in supporting agriculture and industry in Chiba Prefecture. Three sediment cores were collected on 11 August 2011 (Core-1), 30 July 2012 (Core-2) and 24 April 2015 (Core-3) at the same position ($35^\circ 48' 04'' \text{ N}$, $140^\circ 15' 33'' \text{ E}$) near the center of the north lake part using a gravity core sampler (Fig. 1), and the fourth core (Core-4, $35^\circ 48' 06'' \text{ N}$, $140^\circ 15' 42'' \text{ E}$) was collected on 24 April 2015 close to the previous sampling site. These obtained cores had been immediately transported to the land-based lab and stored in a freezer until they were taken out for further pretreatment.

2.2. Analysis of radiocesium and Pu isotopes in sediment samples

After the sediment cores were taken out from the freezer, they were allowed to thaw naturally at room temperature, before each sediment core was sectioned at 1 cm intervals except for the surface layer sample (0-2 cm). After drying the sediment samples at 105°C for 24 h, the activities of ^{137}Cs and ^{134}Cs were determined by gamma-ray spectrometry using germanium detectors (Seiko EG&G, Hatchobori, Japan). An efficiency calibration of the detectors was made with a mixed gamma standard solution (Amersham, QCY- 46; AEA Technology QSA, North Ave., Burlington, MA USA) or a volume radioactivity standard gamma source (MX0333U8, Japan Radioisotopes Association). Reference standard materials

IAEA-156 and IAEA-375 were used for an accuracy check. The counting time for each sample was from 7200 to 300,000 seconds. All the measurement values were corrected to 15 March 2011.

The sample preparation procedures for Pu analysis were described in detail elsewhere (Zheng et al., 2009). Pu was not determined in the Core-4 samples. The flow chart of the analytical procedure for the determination of Pu isotopes is summarized in Figure S1. Briefly, sample digestion was conducted on a hotplate in Teflon beaker after adding 20 mL 10 M HNO₃-1 M HF and 0.57 pg ²⁴²Pu (CRM 130, plutonium spike assay and isotopic standard, New Brunswick Laboratory, USA; the ²⁴²Pu purity was 99.92%), and then the Fe(OH)₃ method was used to co-precipitate Pu isotopes. The two-stage anion-exchange chromatographic method with AG 1X8 and AG MP-1M resins was employed for radiochemical separation and purification of Pu isotopes (Bu et al., 2014b). The final sample was prepared in 4% HNO₃ medium for Pu analysis using sector-field (SF) ICP-MS (Thermo Element XR, Bremen, Germany) combined with an APEX-Q high efficiency sample introduction system (Elemental Scientific Inc., Omaha NE, USA) (Zheng and Yamada, 2006; Zheng, 2015). The analytical method has been previously validated with the analysis of two reference materials, marine sediment (NIST-4357) and soil (IAEA-soil-6), with satisfactory results (Zhang et al., 2010; Wang et al., 2015).

The inventories of $^{239+240}\text{Pu}$ were calculated employing the following equation (Livingston et al., 2001):

$$I_s = \sum_{i=1}^M S_i D_i (U_i - L_i) \quad (1)$$

where I_s is the Pu inventory in the sediment column (Bq/m^2), M is the number of sediment layers (the layer number is counted downward from the surface to the deeper layer), S_i is the Pu concentration in layer i (Bq/kg , dry weight), D_i is the dry bulk density of layer i from the dried samples (kg/m^3), U_i is the upper boundary of layer i (m) and L_i is the lower boundary of layer i (m). Radiocesium inventories of the sediment cores were also estimated based on equation (1) by replacing $^{239+240}\text{Pu}$ with ^{137}Cs activity.

3. Results

3.1. $^{239+240}\text{Pu}$ activities and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in sediment samples of Core-1 to Core -3

The obtained activities of $^{239+240}\text{Pu}$, the atom ratios of $^{240}\text{Pu}/^{239}\text{Pu}$, and the inventories of $^{239+240}\text{Pu}$ are summarized in Table S1 in the supporting information. The $^{239+240}\text{Pu}$ activities for Core-1 ranged from 0.007 to 0.307 Bq/kg with an average of $0.187 \pm 0.021 \text{ Bq/kg}$. For Core-2, the $^{239+240}\text{Pu}$ activities varied between 0.001 and 0.245 Bq/kg , with an average of $0.182 \pm 0.015 \text{ Bq/kg}$. For Core-3, the $^{239+240}\text{Pu}$ activities ranged from 0.015 to 0.285 Bq/kg with an average of $0.197 \pm 0.025 \text{ Bq/kg}$. The vertical distribution of $^{239+240}\text{Pu}$ activities is illustrated in Fig. 2, and the activities of $^{239+240}\text{Pu}$ in the three sediment cores in their surface

and upper layers (approximately 0-16 cm) overall presented no obvious changes, especially for Core-2, with a narrow range of 0.22-0.25 Bq/kg. After 16 cm in the $^{239+240}\text{Pu}$ profiles, the $^{239+240}\text{Pu}$ activities presented a rapid decrease. The inventories of $^{239+240}\text{Pu}$ for Core-1, Core-2 and Core-3 were estimated to be 27.1 ± 1.43 , 27.7 ± 0.67 and 28.1 ± 1.60 Bq/m², respectively. The Pu inventories in the three sediment columns were slightly lower than that of the expected global fallout inventory at 30-40 °N (ca. 42 Bq/m²) (UNSCEAR, 2000). The obtained $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in all the sediment samples of Core-1 to Core-3 were in a range of 0.161-0.210 with an average of 0.198 ± 0.021 (1 σ). More specifically, the ranges of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio were: Core-1, 0.175-0.210; Core-2, 0.161-0.200; Core-3, 0.175-0.205.

3.2. $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios and ^{134}Cs and ^{137}Cs activities in sediment samples of Core-1 to Core-4

The measured activities of ^{134}Cs and ^{137}Cs , the activity ratios of $^{134}\text{Cs}/^{137}\text{Cs}$, and the inventories of ^{134}Cs and ^{137}Cs are presented in Table S1. The vertical distributions of ^{137}Cs activities are shown in Fig. 3. The ^{137}Cs activities in Core-1 ranged from 19 to 879 Bq/kg. The highest activities of radiocesium appeared in the surface and first upper layers (0-3 cm), and a sharp decrease of radiocesium activities after 3 cm depth was observed in the profile. For Core-2, the measured ^{137}Cs activities were in a range of 28-633 Bq/kg (decay-corrected to 15 March 2011). A significant mixing zone from the surface down to 8 cm was observed.

A remarkable decreasing trend of ^{137}Cs activities with depth occurred after the 8 cm depth. For Core-3, the vertical distribution of ^{137}Cs activities was similar to that of Core-2, but the ^{137}Cs activities in upper layers were obvious lower compared with Core-2. The vertical distribution pattern of ^{137}Cs activities for Core-4 was different from the other three cores which may be due to the different sampling site. Surface water of this site was covered by a floating-leaved aquatic plant, *Trapa natans* var. *japonica* from early summer to fall. For examples, the ^{137}Cs activities in Core-4 tended to increase in the layers 0-6 cm and then dramatically decreased for layers deeper than 6 cm. The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios for Core-1 ranged from 0.78 to 1.04 with an average of 0.95 ± 0.10 , and for Core-2 they varied between 0.67 and 1.17, with an average of 0.96 ± 0.11 . For Core-3 they ranged from 0.99 to 1.24 with an average of 1.17 ± 0.09 , and for Core-4, they ranged from 0.95 to 1.52 with an average of 1.14 ± 0.15 . The inventories of ^{137}Cs for Core-1, Core-2, Core-3 and Core-4 were estimated to be 32300 ± 660 , 37500 ± 330 , 30200 ± 570 Bq/m² and 20700 ± 410 Bq/m², respectively. The ^{134}Cs inventories for Core-1, Core-2, Core-3 and Core-4 were calculated to be 30200 ± 590 , 35200 ± 550 , 33200 ± 650 and 23500 ± 380 Bq/m², respectively (decay-corrected to 15 March 2011).

4. Discussion

4.1 Geochemical behaviors and source identification of Pu isotopes in Lake Inba sediments

The activities of $^{239+240}\text{Pu}$ in Japanese lakes (Lake Obuchi, Kizaki, Kugushi, Ichiyanagi, Towada, Tamogi, and Takahoko) have been investigated and overall ranged from 0.02 to 7.00 Bq/kg before the FDNPP accident (Yamamoto et al., 1991; Nagao et al., 1999; Ueda et al., 2004, 2009; Zheng and Yamada, 2005). Clearly, the analytical results for $^{239+240}\text{Pu}$ activities (0.001-0.307 Bq/kg) in Lake Inba were within the range of the expected preexisting level. Among the lakes mentioned above, the highest $^{239+240}\text{Pu}$ activity around 7.00 Bq/kg was observed in Lake Kugushi; this lake was influenced by a large inflow from a river into the lake of fine particulate materials that had adsorbed high amount of radionuclides (Yamamoto et al., 1991).

Generally, for a typical profile of $^{239+240}\text{Pu}$ activities in the lake with stable sedimentary environment, a significant maximum $^{239+240}\text{Pu}$ activity in a sediment core can be observed due to a series of large-scale atmospheric nuclear tests conducted in the early 1960s. For example, remarkable peaks of $^{239+240}\text{Pu}$ activities were observed in different lacustrine cores in Lake Obuchi (Ueda et al., 2004) as well as other lakes in China (Huh et al., 1996; Zheng et al., 2008a, 2008b), and these apparent peaks of $^{239+240}\text{Pu}$ activities corresponded to the period of 1963 and 1964. For Lake Inba, the observed highest $^{239+240}\text{Pu}$ activities at approximately 14 cm, 10 cm and 16 cm in Core-1, Core-2 and Core-3 were 0.307 ± 0.032 , 0.245 ± 0.026 and 0.285 ± 0.034 Bq/kg (Table S1), respectively. However, these highest activities were significantly lower than those in the sediment cores of Lake Obuchi (3.14 ± 0.124 , $5.03 \pm$

0.223, 5.47 ± 0.197 Bq/kg) (Ueda et al., 2004) and Lake Towada (5.24 ± 0.130 Bq/kg) (Nagao et al., 1999). Actually, there are no apparent peaks for $^{239+240}\text{Pu}$ activities in the sediment cores from Lake Inba (Fig. 2). As noted earlier, the activities of $^{239+240}\text{Pu}$ in sediments upper layers (approximately 0-16 cm) exhibited no remarkable changes. The higher $^{239+240}\text{Pu}$ activities in the upper layers in Lake Inba were likely to be caused by great Pu input in the early 1960s due to large-scale nuclear test explosions. Considering the fact that Lake Inba is a typical shallow lake (average depth 1.7 m) with relatively flat topography around the lake, the process of sedimentation was disturbed by physical mixing process, resulting in an even distribution of global fallout sourced $^{239+240}\text{Pu}$ activities in upper layers. Fuller et al. (1999) observed a mixing zone depth of 33 cm from a ^{210}Pb profile obtained in San Francisco Bay, and they pointed out that extensive mixing could result in the persistence of contaminants within the upper layer sediments for many years after deposition. We considered that some kinds of complicated physical and biological processes (such as strong wind mixing and bioturbation) together with sedimentation resulted in redistribution and migration of Pu in Lake Inba. However, identifying the true cause of Pu distribution in the surface and upper layers requires additional investigations such influences as granularity and presence of burrowing organisms.

It is widely acknowledged that the inventory of $^{239+240}\text{Pu}$ plays an important role in geochemical studies as it can reflect the removal efficiency, sources and transportation of Pu

isotopes in particles as well as in water. The estimated $^{239+240}\text{Pu}$ inventories of the cores are presented in Fig. 4. These Pu inventories, like the $^{239+240}\text{Pu}$ activities, were significantly lower than those of Lake Tamogi, Takahoko, Ichiyanagi, Kugushi, Mikata, and Obuchi (170-240 Bq/m²) (Yamamoto et al., 1991; Ueda et al., 2004; Ueda et al., 2009). A possible explanation for this difference was that the previously investigated lakes mentioned above were located at 40-50 °N and they received greater amount of Pu isotopes from global fallout (ca. 58 Bq/m²) (UNSCEAR, 2000). For example, $^{239+240}\text{Pu}$ inventory of Lake Sihailongwan (42°17'N, 126°36'E) in Northeastern China was 62.7 Bq/m² (Wu et al., 2010), and was close to the integrated atmospheric fallout values 58 Bq/m² (UNSCEAR, 2000). In addition, the inflow of Pu isotopes from the catchment areas could contribute to the larger Pu inventories of these lakes such as the Lake Kugushi and Mikata at 30-40 °N (Yamamoto et al., 1991; Ueda et al., 2004; Ueda et al., 2009). For Lake Inba, the dry bulk densities for sediment cores ranged from 0.53 to 1.07 g/cm³, and average values were around 0.75 g/cm³ (Table S1). Obviously, the dry bulk densities for core samples from Lake Inba were quite low compared with those from Lake Obuchi (1.38-2.18 g/cm³ with an average of 1.62 g/cm³) (Zheng and Yamada, 2005), which could partly contribute to low inventories of Pu isotopes. Besides, as mentioned above, the topography of surrounding region of the Lake Inba is relatively flat, thus the sediment input into the lake could be quite limited every year, especially in recent years due to the infiltration facilities (Takashi et al., 2007). We can roughly estimate the sedimentation

253 rate is approximately 0.3 cm/year because most of the input Pu isotopes in the 1960s was
254 mainly in upper layers (0-16 cm) in the past five decades. Therefore, the small sedimentation
255 rate is another reason for the lower Pu inventory in Lake Inba. It is widely acknowledged that
256 precipitation is positively correlated with Pu inventory, while the annual precipitation (2600
257 mm) in the lakes (Lake Kugushi and Mikata) situated Fukui Prefecture (Sugimoto et al., 2015)
258 was almost twice higher than that of Lake Inba (1400 mm) in Chiba Prefecture (Okuda et al.,
259 2005), which may also result in lower inventory in Lake Inba.

260 Besides global fallout Pu, another potential source of Pu contamination in the study area
261 was tropospheric fallout from the nuclear weapons tests conducted in the Pacific Proving
262 Grounds (PPG) during 1946-1958 in the North Pacific. The PPG-sourced Pu bearing a high
263 $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of 0.30-0.36 has been widely observed in the Northwest Pacific marine
264 environment (sediments and seawaters) surrounding Japan (Zheng and Yamada, 2004).
265 Spatial distributions of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in surface marine sediments in the western
266 North Pacific are summarized in Fig. 5. We noted that values of a majority of the $^{240}\text{Pu}/^{239}\text{Pu}$
267 atom ratios (0.140-0.293) in marine sediments around Japan were greater than that of global
268 fallout and lower than that of the PPG source (Table 1), which indicated that Pu isotopes
269 primarily originated from global fallout and the PPG close-in fallout (Buesseler, 1997; Lee et
270 al., 1998). It has been considered that the oceanic current transport of Pu from the PPG
271 source was responsible for the wide presence of the PPG source Pu signature in the

Northwest Pacific as well as its marginal seas, however, the early direct PPG close-in fallout deposition has also been proposed (Wang and Yamada, 2005), and is still an issue of debate (Liu et al., 2011). If the direct PPG close-in fallout Pu reached the main Japanese Island, the PPG-sourced Pu with its high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio could have deposited in the deeper layer lacustrine sediments. As Fig. 6 demonstrates, the values for $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios (0.161-0.210) in the sediment samples were all within the range of global fallout (0.18 ± 0.014) (Kelley et al., 1999), which indicated that the early tropospheric fallout Pu isotopes from the PPG were not transported to the study regions (36 °N), therefore we have been able to narrow further the scope of the PPG-derived Pu isotopes in terrestrial environments. However, more extensive studies are still required on the lacustrine sediment record of Pu atomic ratios and activities in the southern part of Japan to more accurate understanding of the deposition of the PPG-derived Pu isotopes by early direct tropospheric fallout in the Northwest Pacific.

According to the obtained $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios, release of Pu isotopes from the FDNPP accident to sediment samples in Inba Lake was negligible. As mentioned before, the total amount of $^{239+240}\text{Pu}$ derived from the FDNPP accident was estimated to be from 1.0 to 6.4×10^9 Bq (Yamamoto et al., 2012; Zheng et al., 2012b, 2013). If we assume that all of the Fukushima-derived Pu (6.4×10^9 Bq) was deposited evenly in the area within a radius of 200 km from the FDNPP site and these Pu isotopes were soon incorporated with particles sinking

to the bottom of Lake Inba, the estimated excess $^{239+240}\text{Pu}$ inventory could be approximately 0.05 Bq/m² and this trace amount of Pu isotopes compared to $^{239+240}\text{Pu}$ inventory (approximately 27.6 Bq/m²) in the sediment column would be negligible.

4.2 Source term of radiocesium and its accumulation in the sediment cores

The Fukushima-source radiocesium was characterized by a $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio of 1 on 15 March 2011 (Chino et al., 2011; Ishii et al., 2013). In this study, the observed $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios of approximately 1 in sediment samples were consistent with reported values, which indicated that these sediment samples from Lake Inba were contaminated with radioactive Cs from the FDNPP accident. We summarized published studies regarding the activities of ^{137}Cs in sediments of different lakes and rivers in Japan before the FDNPP accident (Nagao et al., 1999; Mizugaki et al., 2006; Ueda et al., 2009; Kanai, 2011), and the activities of ^{137}Cs in sediment samples were generally less than 130 Bq/kg (decay-corrected to 15 March 2011). Clearly, the detected activities of ^{137}Cs in sediments of Lake Inba were significantly higher (6-7 times) than the background values in lake sediments, which further demonstrated that the radiocesium contamination was caused by the accident.

Fukushima and Arai (2014) reported that the ^{137}Cs activities in surface sediments of Lake Inba were approximately 800 Bq/kg (decay-corrected to 15 March 2011). Subsequently, the

activities of ^{137}Cs in surface sediments exhibited decreasing trends during 2011-2014 (MOE, 2011-2014) (Table S2 and Figure S2). Similarly, the same trends have been observed in our results of radiocesium activities in surface sediments (0-2 cm) (Fig. 3). In addition, the ^{137}Cs inventories of surface layer sediments (0-2 cm) for Core-1, Core-2, and Core-3 accounted for about 34%, 17%, and 15% of total inventories, respectively, and they also showed a decreasing tendency; two possible mechanisms could account for these findings. (1) The effect of hydraulic flushing resulted in the radiocesium being removed out of the lake (due to Cs ion removal from the sediments); (2) The reduced radiocesium amount in the surface layer migrated downward in the sediment column, and still remained in the lake. In order to find the reasonable explanation for the reduced radiocesium amount, we investigated the vertical distributions of ^{137}Cs activities in the sediment columns. Compared with temporal variations of ^{137}Cs profiles (Core-1, Core-2, Core-3 and Core-4) (Fig. 3), although the ^{137}Cs activities in surface sediments showed a decreasing tendency, the activities of ^{137}Cs in the subsurface layer (3-8 cm) tended to increase. Hence, the occurrence of ^{137}Cs downward migration from the surface layer (0-2 cm) to deeper layers (around 8 cm) could be clearly observed. We concluded that the reduced radiocesium amount in the surface layer was caused by downward migration. Recently, Mikami et al (2015) reported that the inventory of ^{137}Cs was approximately 10-60 kBq/m² around Lake Inba. Overall, our results for inventories of radiocesium (Table S1) displayed a relatively good agreement with the reported values

329 ([Mikami et al., 2015](#)), suggesting that most of the FDNPP-derived radiocesium was
330 accumulated in the sediment cores instead of being removed out of the lake by hydraulic
331 flushing.

332 Comparing profiles of ^{137}Cs with $^{239+240}\text{Pu}$, we saw an apparent difference between the
333 vertical distributions of these two radionuclides, which resulted from the difference in the
334 amounts of the FDNPP-released radiocesium and Pu inputs into the lake: deposition input of
335 the FDNPP-released radiocesium was significant, but the extra Pu that originated from the
336 accident could be ignored. Another reason that could account for the difference was that most
337 of the global fallout Pu (deposited in the 1960s) has been present in the lake for about 50
338 years, while input of the FDNPP-derived radiocesium has only occurred since 2011. In spite
339 of radiocesium and Pu isotopes having different chemical properties, high correlations were
340 obtained in lake and marine sediments, indicating that these two radionuclides can provide
341 the similar information about the sedimentation process ([Zheng et al., 2008](#); [Pan et al., 2011](#)).
342 Therefore, the profile of Pu isotopes can be applied as an indicator to predict the geochemical
343 behavior of radiocesium in Lake Inba in the future. Based on this view point, we consider that
344 the FDNPP-originated radiocesium might be mixing continuously as it migrates downward,
345 namely, the activity of radiocesium within the mixing zone is likely to lead to uniform in
346 upper layers in the future.

347 In order to better compare the ^{137}Cs inventories, we collected data regarding ^{137}Cs
348 deposition density in Chiba Prefecture after the accident (NRA, 2012). The spatial
349 distribution of ^{137}Cs deposition density in Chiba Prefecture is presented in Figure S3. The
350 total deposition density of ^{137}Cs was around 27 kBq/m² in the regions surrounding Lake Inba.
351 Our obtained ^{137}Cs inventories in the sediment cores were comparable with this value,
352 indicating that most of the radiocesium in Lake Inba was mainly derived from atmospheric
353 deposition, and the addition of particle-adsorbed Cs from surrounding areas of Lake Inba was
354 not significant. Therefore, we concluded that the input of radiocesium was greater than that of
355 output in Lake Inba, and finally the Fukushima-derived radiocesium was accumulated in the
356 sediments. However, the presently obtained inventories of ^{137}Cs were almost 1-2 orders of
357 magnitude lower than those of lakes (206-360 kBq/m²) closer (40-50 km) to FDNPP site due
358 to much the heavier contamination there (Yoshimura et al., 2014). It has been demonstrated
359 that trapping efficiency or sediment focusing can be calculated as the ratio of the measured
360 radioisotope inventory to the inventory expected due to direct input in the study region (Aller
361 et al., 1980; Davison et al., 1993). The trapping efficiencies were evaluated for several ponds
362 around the FDNPP site, and lower trapping efficiencies (less than 100%) suggested some of
363 the initial radiocesium inputs were removed through hydraulic flushing due to the coarser
364 sediment (Yoshimura et al., 2014). As for Lake Inba, the direct input of ^{137}Cs was calculated
365 by summing up this radionuclide from the FDNPP accident (27 kBq/m²) (NRA, 2012) and

global fallout (1.34 kBq/m^2) (UNSCEAR, 2000). The estimated trapping efficiencies from ^{137}Cs for Core-1 and Core-2 of around $110 \pm 5 \%$ were indicative of net accumulation. It has been documented that the ^{137}Cs accumulation in sediments predominantly occurred by adsorbing onto sediment and sedimentation with suspended and finer particles contained clay minerals (Santschi et al., 1990; Davison et al., 1993; Nagao et al., 1999; Smith et al., 2000; Korobova et al., 2007). Therefore, the remaining radiocesium could be adsorbed onto suspended and finer particles. The ignition loss in sediment samples of Lake Inba ranged from 5% to 20% with an average of 11% (Table S1), suggesting that the sediments were relatively rich in organic substances. The abundance of organic substances in Lake Inba sediments would contribute significantly to radiocesium accumulation because of the effective adsorption of radiocesium by the organic substances in the lake water (Nagao et al., 1999). However, in order to more accurately and systematically quantify the remaining radiocesium budget in the lake system, ^{137}Cs concentration information with regards to fish, plankton and lake water are necessary.

5. Conclusions

In the present work, we investigated the vertical profiles of Pu and radiocesium isotopes in sediment cores from Lake Inba after the FDNPP accident. The obtained $^{239+240}\text{Pu}$ activities in sediment samples were comparable with the background level before the accident. The

$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in these sediment samples were within the range of global fallout.

Thus Pu from the PPG source through the early tropospheric fallout was not delivered to the

regions above 36 °N and Pu contamination caused by the FDNPP accident was negligible.

Additionally, mixing layer can be observed in the upper layers in Lake Inba based on the

profiles of $^{239+240}\text{Pu}$ and ^{137}Cs . More importantly, from this we thought the vertical profiles of

Pu isotopes could be applied to predict the geochemical behavior of ^{137}Cs in the Lake Inba in

the future. The observed $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios of upper sediment layers were around 1,

indicating that radiocesium originated from the accident. The calculated inventories of

radiocesium in the sediment cores suggested that the input of radiocesium was greater than

that of output in this lake.

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404

405 **References**

406 Aller, R. C., Benninger, L. K., Cochran, J. K., 1980. Tracking particle associated processes in
407 nearshore environments by use of $^{234}\text{Th}/^{238}\text{U}$ disequilibrium. *Earth Planet. Sci. Lett.* 47,
408 161-175.

409 Amano, K., Nakanishi, S., 2008. Water quality change in eutrophic lakes due to diversion and
410 water supply from nearby rivers, case for Inba marsh. *Proceedings of Hydraulic*
411 *Engineering.* 52, 1267-1272.

412 Aoyama, M., Tsumune, D., Hamajima, Y., 2013. Distribution of ^{137}Cs and ^{134}Cs in the North
413 Pacific Ocean: impacts of the TEPCO Fukushima-Daiichi NPP accident. *J. Radioanal.*
414 *Nucl. Chem.* 296, 535-539.

415 Bu, W., Fukuda, M., Zheng, J., Aono, T., Ishimaru, T., Kanda, J., Yamada, M., 2014a.
416 Release of Pu isotopes from the Fukushima Daiichi Nuclear Power Plant accident to the
417 marine environment was negligible. *Environ. Sci. Technol.* 48(16), 9070-9078.

418 Bu, W., Zheng, J., Guo, Q. J., Aono, T., Tazoe, H., Tagami, K., Uchida, S., Yamada, M.,
419 2014b. A method measurement of ^{239}Pu , ^{240}Pu and ^{241}Pu in high U content marine
420 sediments by sector field ICP-MS and its application to Fukushima sediment samples.
421 *Environ. Sci. Technol.* 48(1), 534-541.

422 Buesseler, K. O., 1997. The isotopic signature of fallout plutonium in the North Pacific. J.
 423 Environ. Radioact. 36(1), 69-83.

424 Buesseler, K., Aoyama, M., Fukasawa, M., 2011. Impacts of the Fukushima nuclear power
 425 plants on marine radioactivity. Environ. Sci. Technol. 45(23), 9931-9935.

426 Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G., Yamazawa, H. 2011.
 427 Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from
 428 the Fukushima Daiichi nuclear power plant into the atmosphere. J. Nucl. Sci. Technol.
 429 48(7), 1129-1134.

430 Davison, W., Hilton, J., Hamilton-Taylor, J., Kelly, M., Livens, F., Rigg, E., Singleton, D.L.,
 431 1993. The transport of Chernobyl-derived radiocaesium through two freshwater lakes in
 432 Cumbria, UK. J. Environ. Radioact. 19(2), 125-153.

433 Fukushima, T., Arai, H., 2014. Radiocesium contamination of lake sediments and fish
 434 following the Fukushima nuclear accident and their partition coefficient. Inland Waters.
 435 4(2), 204-214.

436 Fuller, C. C., Van Geen, A., Baskaran, M., Anima, R., 1999. Sediment chronology in San
 437 Francisco Bay, California, defined by ^{210}Pb , ^{234}Th , ^{137}Cs , and $^{239,240}\text{Pu}$. Mar. Chem. 64(1),
 438 7-27.

439 Hirose, K., 2012. Fukushima Daiichi nuclear power plant accident: summary of regional
 440 radioactive deposition monitoring results. J. Environ. Radioact. 111, 13-17.

441 Huh, C. A., Chu, K. S., Wei, C. L., Liew, P. M., 1996. Lead-210 and plutonium fallout in
 442 Taiwan as recorded at a subalpine lake. *J. Asian. Earth. Sci.* 14, 373-376.

443 IAEA, 2015. The Fukushima Daiichi accident. Technical Volume 1/5 Description and
 444 Context of the Accident. International Atomic Energy Agency, Vienna, Austria.

445 Ishii, N., Tagami, K., Takata, H., Fujita, K., Kawaguchi, I., Watanabe, Y., Uchida, S., 2013.
 446 Deposition in Chiba Prefecture, Japan, of Fukushima Daiichi nuclear power plant fallout.
 447 *Health. Phys.* 104(2), 189-194.

448 Kanai, Y., 2011. Characterization of ^{210}Pb and ^{137}Cs radionuclides in sediment from Lake
 449 Shinji, Shimane Prefecture, western Japan. *Appl. Radiat. Isotopes.* 69(2), 455-462.

450 Kelley, J. M., Bond, L. A., Beasley, T. M., 1999. Global distribution of Pu isotopes and ^{237}Np .
 451 *Sci. Total Environ.* 237, 483-500.

452 Ketterer, M. E., Hafer, K. M., Mietelski, J. W., 2004. Resolving Chernobyl vs. global fallout
 453 contributions in soil from Poland using plutonium atom ratios measured by inductively
 454 coupled plasma mass spectrometry. *J. Environ. Radioact.* 73, 183-201.

455 Korobova, E. M., Chizhikova, N. P., 2007. Distribution and mobility of radiocesium in
 456 relation to the clay fraction mineralogy and soil properties in the Iput River floodplain.
 457 *Eurasian Soil Sci.* 40, 1062-1075.

458 Lee, M. H., Lee, C. W., Moon, D. S., Kim, K. H., Boo, B. H., 1998. Distribution and
 459 inventory of fallout Pu and Cs in the sediment of the East Sea of Korea. J. Environ.
 460 Radioact. 41, 99-110.

461 Liu, Z. Y., Zheng, J., Pan, S. M., Dong, W., Yamada, M., Aono, T., Guo, Q. J., 2011. Pu and
 462 ¹³⁷Cs in the Yangtze River Estuary sediments: distribution and source identification.
 463 Environ. Sci. Technol. 45, 1805-1811.

464 Livingston, H. D., Povinec, P. P., Ito, T., Togawa, O., 2001. The behaviour of plutonium in
 465 the Pacific Ocean, in: Kudo, A. (Ed), Plutonium in the Environment, Elsevier Science Ltd,
 466 Oxford, UK, pp. 267-292.

467 METI (Ministry of Economy, Trade and Industry, Japan). Data on the Amount of released
 468 Radioactive Materials, 2011. (<http://www.meti.go.jp/press/2011/10/20111020001/20111020001.pdf>).
 469 020001. pdf).

470 Mikami, S., Maeyama, T., Hoshide, Y., Sakamoto, R., Sato, S., Okuda, N. Demongeot, S.,
 471 Gurriarang, R., Uwaminoh, Y., Katoi, H., Fujiwaraj, M., Satok, T., Takemiyal, H., Saitoa,
 472 K., 2015. Spatial distributions of radionuclides deposited onto ground soil around the
 473 Fukushima Dai-ichi Nuclear Power Plant and their temporal change until December 2012.
 474 J. Environ. Radioact. 139, 320-343.

475 Miyake, Y., Matsuzaki, H., Fujiwara, T., Saito, T., Yamagata, T., Honda, M., Muramatsu, Y.,
 476 2012. Isotopic ratio of radioactive iodine ($^{129}\text{I}/^{131}\text{I}$) released from Fukushima Daiichi NPP
 477 accident. *Geochem. J.* 46(4), 327-333.

478 Mizugaki, S., Nakamura, F., Araya, T., 2006. Using dendrogeomorphology and ^{137}Cs and
 479 ^{210}Pb radiochronology to estimate recent changes in sedimentation rates in Kushiro Mire,
 480 Northern Japan, resulting from land use change and river channelization. *Catena*. 68(1),
 481 25-40.

482 MOE (Ministry of the Environment, Japan), 2011-2014. Available from: [http://www.env.go.](http://www.env.go.jp/jishin/monitoring/results_r-pw)
 483 [jp/jishin/monitoring/results_r-pw](http://www.env.go.jp/jishin/monitoring/results_r-pw).

484 Morino, Y., Ohara, T., Nishizawa, M. 2011. Atmospheric behavior, deposition, and budget of
 485 radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011,
 486 *Geophys. Res. Lett.* 38 (7). <http://dx.doi.org/10.1029/2011GL048689>.

487 Muramatsu, Y., Rühm, W., Yoshida, S., Tagami, K., Uchida, S., Wirth, E., 2000.
 488 Concentrations of ^{239}Pu and ^{240}Pu and their isotopic ratios determined by ICP-MS in soils
 489 collected from the Chernobyl 30 km zone. *Environ. Sci. Technol.* 34(14), 2913-2917.

490 Nagao, S., Matsunaga, T., Muraoka, S., 1999. Geochemical association of ^{137}Cs and $^{239,240}\text{Pu}$
 491 in the oligotrophic and mesotrophic lake sediments. *J. Radioanal. Nucl. Chem.* 239(3),
 492 555-559.

493 NRA (Nuclear Regulation Authority, Japan), 2012. Available from:
 494 <http://radioactivity.nsr.go.jp/ja/contents/7000/6212/view>.

495 Ochiai, S., Nagao, S., Yamamoto, M., Itono, T., Kashiwaya, K., Fukui, K., Iida, H., 2013.
 496 Deposition records in lake sediments in western Japan of radioactive Cs from the
 497 Fukushima Dai-ichi nuclear power plant accident. *Appl. Radiat. Isotopes*. 81, 366-370.

498 Okuda, T., Iwase, T., Ueda, H., Suda, Y., Tanaka, S., Dokiya, Y., Hosoe, M., 2005.
 499 Long-term trend of chemical constituents in precipitation in Tokyo metropolitan area,
 500 Japan, from 1990 to 2002. *Sci. Total. Environ.* 339(1), 127-141.

501 Pan, S. M., Tims, S. G., Liu, X. Y., Fifield, L. K., 2011. ^{137}Cs , $^{239+240}\text{Pu}$ concentrations and
 502 the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in a sediment core from the sub-aqueous delta of Yangtze River
 503 estuary. *J. Environ. Radioact.* 102(10), 930-936.

504 Saito, K., Tanihata, I., Fujiwara, M., Saito, T., Shimoura, S., Otsuka, T., Onda, Y., Hoshi, M.,
 505 Ikeuchi, Y., Takahashi, F., Kinouchi, N., Saegusa, J., Seki, A., Takemiya, H., Shibata, T.,
 506 2015. Detailed deposition density maps constructed by large-scale soil sampling for
 507 gamma-ray emitting radioactive nuclides from the Fukushima Dai-ichi Nuclear Power
 508 Plant accident. *J. Environ. Radioact.*, 139, 308-319.

509 Santschi, P. H., Bollhalder, S., Zingg, S., Lück, A., Farrenkothen, K., 1990. The self-cleaning
 510 capacity of surface waters after radioactive fallout. Evidence from European waters after
 511 Chernobyl, 1986-1988. *Environ. Sci. Technol.* 24(4), 519-527.

512 Shinonaga, T., Steier, P., Lagos, M., Ohkura, T., 2014. Airborne plutonium and non-natural
 513 uranium from the Fukushima DNPP found at 120 km distance a few days after reactor
 514 hydrogen explosions. *Environ. Sci. Technol.* 48(7), 3808-3814.

515 Smith, J. T., Comans, R. N. J., Ireland, D. G., Nolan, L., Hilton, J., 2000. Experimental and in
 516 situ study of radiocaesium transfer across the sediment–water interface and mobility in
 517 lake sediments. *Appl. Geochem.* 15(6), 833-848.

518 Sugimoto, R., Tomoko, T., 2015. Seasonal and annual fluxes of atmospheric nitrogen
 519 deposition and riverine nitrogen export in two adjacent contrasting rivers in central Japan
 520 facing the Sea of Japan. *J. Hydrol. Regional Studies*. [http://dx.doi.org/10.1016/j.ejrh.201](http://dx.doi.org/10.1016/j.ejrh.2015.11.019)
 521 [5.11.019](http://dx.doi.org/10.1016/j.ejrh.2015.11.019).

522 Tagami, K., Uchida, S., 2015. Effective half-lives of ^{137}Cs in giant butterbur and field
 523 horsetail, and the distribution differences of potassium and ^{137}Cs in aboveground tissue
 524 parts. *J. Environ. Radioact.* 141, 138-145.

525 TEPCO (Tokyo Electric Power Company), 2012. <http://www.tepco.co.jp/en/press/corp-com>
 526 [/release/11040815-e.html](http://www.tepco.co.jp/en/press/corp-com/release/11040815-e.html).

527 Ueda, S., Ohtsuka, Y., Kondo, K., 2004. Inventories of $^{239+240}\text{Pu}$, ^{137}Cs , and excess ^{210}Pb in
 528 sediment cores from brackish Lake Obuchi, Rokkasho Village, Japan. *J. Radioanal. Nucl.*
 529 *Chem.* 261(2), 277-282.

530 Ueda, S., Ohtsuka, Y., Kondo, K., Hisamatsu, S. I., 2009. Inventories of $^{239+240}\text{Pu}$, ^{137}Cs , and
 531 excess ^{210}Pb in sediments from freshwater and brackish lakes in Rokkasho, Japan,
 532 adjacent to a spent nuclear fuel reprocessing plant. J. Environ. Radioact. 100(10),
 533 835-840.

534 UNSCEAR, 1982. Ionizing radiation: sources and biological effects, United Nations
 535 Scientific Committee on the Effects of Atomic Radiation Exposures to the Public from
 536 Man-made Sources of Radiation, United Nations, New York.

537 UNSCEAR, 2000. Sources and Effects of Ionizing Radiation, Vol. 1, United Nations
 538 Scientific Committee on the Effects of Atomic Radiation Exposures to the Public from
 539 Man-made Sources of Radiation, United Nations, New York.

540 Wang, Z., Yamada, M., 2005. Pu activities and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in sediment cores
 541 from the East China Sea and Okinawa Trough: sources and inventories. Earth Planet. Sci.
 542 Lett. 233, 441-453.

543 Wang, Z., Yang, G., Zheng, J., Cao, L., Yu, H., Zhu, Y., Uchida, S., 2015. Effect of ashing
 544 temperature on accurate determination of plutonium in soil samples. Anal. Chem. 87(11),
 545 5511-5515.

546 Wu, F., Zheng, J., Liao, H., Yamada, M., 2010. Vertical distributions of plutonium and ^{137}Cs
 547 in lacustrine sediments in northwestern China: quantifying sediment accumulation rates
 548 and source identifications. Environ. Sci. Technol., 44(8): 2911-2917.

549 Yamada, S., Kitamura, A., Kurikami, H., Yamaguchi, M., Malins, A., Machida, M., 2015.
 550 Sediment and ^{137}Cs transport and accumulation in the Ogaki Dam of eastern Fukushima.
 551 Environ. Res. Lett. 10(1), 014013.

552 Yamamoto, M., Sakaguchi, A., Ochiai, S., Takada, T., Hamataka, K., Murakami, T., Nagao,
 553 S., 2014. Isotopic Pu, Am and Cm signatures in environmental samples contaminated by
 554 the Fukushima Dai-ichi Nuclear Power Plant accident. J. Environ. Radioact. 132, 31-46.

555 Yamamoto, M., Takada, T., Nagao, S., Koike, T., Shimada, K., Hoshi, M., Zhumgdilov, K.,
 556 Shima, T., Fukuoka, M., Imanaka, T., Endo, S., Sakaguchi, A., Kimura, S., 2012. An
 557 early survey of the radioactive contamination of soil due to the Fukushima Dai-ichi
 558 Nuclear Power Plant accident with emphasis on plutonium analysis, Geochem. J. 46,
 559 341-353.

560 Yamamoto, M., Yamauchi, Y., Chatani, K., Igarashi, S., Komura, K., Ueno, K., Sakanoue,
 561 M., 1991. Distribution of global fallout ^{237}Np , Pu isotopes, and ^{241}Am in lake and sea
 562 sediments. J. Radioanal. Nucl. Chem. 147(1), 165-176.

563 Yoshida, N., Kanda, J., 2012. Tracking the Fukushima radionuclides. Science. 336,
 564 1115-1116.

565 Yoshimura, K., Onda, Y., Fukushima, T., 2014. Sediment particle size and initial radiocesium
 566 accumulation in ponds following the Fukushima DNPP accident. Sci. Rep. 2, 304 DOI:
 567 10.1038/srep04514.

568 Yuasa, T., Furukawa, I., Masuoka, Y., Musiake, K, 2007. Chinese Hydraulic Engineering
569 Society 2007 Annual Meeting.

570 Zhang, Y., Zheng, J., Yamada, M., Wu, F., Igarashi, Y., Hirose, K., 2010. Characterization of
571 Pu concentration and its isotopic composition in a reference fallout material. *Sci. Total*
572 *Environ.* 408(5), 1139-1144.

573 Zheng, J., 2015. Evaluation of a new sector-field ICP-MS with Jet Interface for ultra-trace
574 determination of Pu isotopes: from femtogram to attogram levels. *J. Nucl. Radioanal. Sci.*
575 15(1), 7-13.

576 Zheng, J., Yamada, M., 2004. Sediment core record of global fallout and Bikini close-in
577 fallout Pu in Sagami Bay, western northwest Pacific margin. *Environ. Sci. Technol.*
578 38(13), 3498-3504.

579 Zheng, J., Yamada, M., 2005. Investigating Pu and U isotopic compositions in sediments: a
580 case study in Lake Obuchi, Rokkasho Village, Japan using sector-field ICP-MS and
581 ICP-QMS. *J. Environ. Monit.* 7(8), 792-797.

582 Zheng, J., Yamada, M., 2006. Determination of U isotope ratios in sediments using ICP-QMS
583 after sample cleanup with anion-exchange and extraction chromatography. *Talanta.* 68(3),
584 932-939.

585 Zheng, J., Aono, T., Uchida, S., Zhang, J., and Honda, M. C., 2012a. Distribution of Pu
 586 isotopes in marine sediments in the Pacific 30 km off Fukushima after the Fukushima
 587 Daiichi nuclear power plant accident. *Geochem. J.* 46, 361-369.

588 Zheng, J., Liao, H., Wu, F., Yamada, M., Fu, P., Liu, C., Wan, G., 2008a. Vertical
 589 distributions of $^{239+240}\text{Pu}$ activity and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in sediment core of Lake
 590 Chenghai, SW China. *J. Radioanal. Nucl. Chem.* 275(1), 37-42.

591 Zheng, J., Tagami, K., Watanabe, S., Uchida, S., Aono, T., Ishii, N., Yoshida, S., Kubota, Y.,
 592 Fuma, S., Ihara, S., 2012b. Isotopic evidence of plutonium release into the environment
 593 from the Fukushima DNPP accident. *Sci. Rep.* 2, 304 DOI: 10.1038/srep00304.

594 Zheng, J., Tagami, K., Uchida, S., 2013. Release of plutonium isotopes into the environment
 595 from the Fukushima Daiichi nuclear power plant accident: what is known and what needs
 596 to be known. *Environ. Sci. Technol.* 47, 9584-9595.

597 Zheng, J., Wu, F., Yamada, M., Liao, H., Liu, C., Wan, G., 2008b. Global fallout Pu recorded
 598 in lacustrine sediments in Lake Hongfeng, SW China. *Environ. Pollut.* 152(2), 314-321.

599 Zheng, J., Yamada, M., Wu, F., Liao, H., 2009. Characterization of Pu concentration and its
 600 isotopic composition in soils of Gansu in northwestern China. *J. Environ. Radioact.*
 601 100(1), 71-75.

Table 1. Summary of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio and $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio of various sources in the environment.

Sources	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio	$^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio	Reference
Global fallout	0.18 ± 0.014 (30-71°N)	0.25	UNSCEAR,1982; Kelley et al., 1999
FDNPP accident	0.303-0.330	1.07-2.89	TEPCO, 2012; Zheng et al., 2012b
Pacific Proving Grounds	0.3		Buesseler, 1997
Chernobyl accident	0.4	0.49-0.56	Muramatsu et al., 2000; Ketterer et al., 2004

619 **Figure captions**

620 **Fig. 1** Map of sampling sites in the north part of Lake Inba.

621 **Fig. 2** Vertical distribution of $^{239+240}\text{Pu}$ activities in the three sediment cores.

622 **Fig. 3** Vertical distributions and temporal variations of ^{137}Cs activities in the four sediment
623 cores.

624 **Fig. 4** Comparison of $^{239+240}\text{Pu}$ inventories obtained in sediment cores and other lakes in
625 Japan. Inventory data for Lake Kugushi and Mikata were adopted from Yamamoto et
626 al. (1991), Lake Obuchi from Ueda et al. (2004), and Lake Ichiyanagi, Tamogi and
627 Takahoko from Ueda et al. (2009).

628 **Fig. 5** Spatial distribution of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in surface sediments in the Northwest
629 Pacific during the 1990s-2010s (Zheng et al., 2012a, and more references cited
630 therein).

631 **Fig. 6** $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in the sediment samples and Fukushima environment samples.
632 The closed blue, red and green circles represent sediments in this study; the closed
633 pink circles (soil and litter samples, cited from Zheng et al. 2012b), closed purple
634 circles (soil and black substances adopted from Yamamoto et al. 2014) and closed
635 orange circles (aerosol samples adopted from Shinonaga et al. 2014) represent the
636 FDNPP-sourced data.

637

Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.