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Determination of plutonium isotopes in marine sediments off the Fukushima coast following the Fukushima Dai-ichi Nuclear Power Plant accident

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Abstract

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident led to the release of large amounts of radionuclides into the atmosphere as well as direct discharges into the sea. In contrast to the intensive studies on the distribution of the released high volatility fission products, such as ^{131}I, ^{134}Cs and ^{137}Cs, similar studies of the actinides, especially the Pu isotopes, are limited. To obtain the vertical distribution of Pu isotopes in marine sediments and to better assess the possible contamination of Pu from the FDNPP accident in the marine environment, we determined the activities of $^{239+240}Pu$ and ^{241}Pu as well as the atom ratios of $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ in sediment core samples collected in the western North Pacific off Fukushima from July 2011 to July 2012. We also measured surface sediment samples collected from seven Japanese estuaries before the FNDPP accident to establish the comprehensive background baseline data. The observed results of both the Pu activities and the Pu atom ratios for the sediments in the western North Pacific were comparable to the baseline data, suggesting that the FDNPP accident did not cause detectable Pu contamination to the studied regions prior to the sampling time. The Pu isotopes in the western North Pacific 30 km off Fukushima coast originated from global fallout and Pacific Proving Ground close-in fallout.

1 Introduction

On 11 March 2011, a magnitude 9.0 earthquake, centered in the northwest Pacific about 130 km off the northeast coast of Japan, and the ensuing gigantic tsunami caused severe damage to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). The cooling systems of some of the nuclear reactor units failed, resulting in hydrogen explosions in the reactor buildings and venting of gases, which caused large releases of radionuclides into the atmosphere. For example, the total released amount of ^{137}Cs to the atmosphere from the FDNPP reactors has been estimated to be in the range of

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9.9–36.6 PBq (Chino et al., 2011; Morino et al., 2011; Stohl et al., 2012). More than 70 % of the released radionuclides were deposited over the North Pacific (Yoshida and Kanda, 2012). In addition, highly contaminated water with large amounts of radionuclides, originating from desperate attempts to prevent reactor cores meltdowns by injecting water into the reactor units, was directly leaked or discharged into the North Pacific Ocean (Buesseler et al., 2011; Inoue et al., 2012; Tsumune et al., 2012).

Intensive studies on the high volatility fission products released into the ocean, such as ^{131}I, ^{134}Cs and ^{137}Cs, were carried out after the FDNPP accident, and data on the concentration and distribution of these products were immediately collected (Aoyama et al., 2013; Buesseler et al., 2011; Honda et al., 2012; Inoue et al., 2012). However, similar studies focusing on the possible released actinides, especially Pu isotopes, are limited. Pu isotopes are characterized by high chemical toxicity, radiotoxicity and long half-lives, and they have attracted much scientific and public concern. Investigating the impact of the FDNPP accident on the distribution of long-half live radionuclides in the environment is important for the long-term dose assessments (Yoshida and Kanda, 2012). In addition, as Pu isotopes are produced by the initial neutron capture reaction in ^{238}U in the nuclear reactors and directly used as the component of MOX fuel in the FDNPP unit 3 reactor (Burns et al., 2012), accurate determination of Pu isotopes in the environmental materials may provide important information to understand the reactors damages.

Abnormal atom ratios of ^{240}Pu / ^{239}Pu (> 0.3) and activity ratios of ^{241}Pu / $^{239+240}$Pu (> 100) were reported in the surface soil and litter samples in the 20–30 km zone around the FDNPP, providing evidence for the atmospheric release of Pu into the terrestrial environment (Zheng et al., 2012a). Imanaka et al. (2012) and Yamamoto et al. (2012) also found ^{238}Pu / $^{239+240}$Pu activities ratios, higher than the global fallout value, for soil samples collected in Iitate Village and Okuma Town, at distances of 25–45 km from the plant. In the marine environment, we investigated the distribution of Pu isotopes in surface marine sediments in the Pacific 30 km off the FDNPP that were collected several months after the accident (Zheng et al., 2012b), and observed

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no significant variation in $^{239+240}$Pu activity and ^{240}Pu / ^{239}Pu atom ratio compared with previously reported values in marine sediments in the western North Pacific. Meanwhile, Sakaguchi et al. (2012) found that Pu concentration in seawater sampled from the Pacific Ocean 50 km from the FDNPP site showed no extra component from the accident. However, the possible long-term Pu contamination in the marine environment remains unknown as no information is currently available on the Pu isotopes in the released radioactive liquids and in the FDNPP near-coastal (within 30 km) marine environment. Pu isotopes in the western North Pacific off Japan can be transported rapidly by oceanic currents, e.g. the Oyashio Current and the Kuroshio Current, and undergo advection and mixing (Buesseler et al., 2012; Lee et al., 2005; Zheng and Yamada, 2005). Thus, the possible Pu contamination from the accident needs further investigation before reaching a reliable conclusion.

Investigating the concentration of radionuclides in the marine sediments is important for the radiobiological assessment in the marine environment as the sediments are a possible source of continued contamination for the marine biota (Buesseler, 2012). The radiocesium (^{134}Cs and ^{137}Cs) originating from the accident was detected in the sinking particles collected from the deep sea (water depth 4810 m) in the western North Pacific, one month after the accident, suggesting the quick transportation of radionuclides to the deep sea via atmospheric deposition (Honda et al., 2013). Otasaka and Kobayashi (2012) observed that dissolved radiocesium from the FDNPP accident was advected southward where it deposited onto sediments along the Ibaraki coast early in the accident. Pu isotopes are more particle-reactive than Cs and the sediment-water distribution coefficient (K_d value) of Pu is two orders of magnitude higher than that of Cs (IAEA, 2004). In mid-April 1986, by a coincidence of timing, IAEA-MEL scientists moored automated time-series sediment traps in the Ligurian Sea, and they found that in one month after the accident more than 50 % of the total $^{239+240}$Pu inventory originating from Chernobyl and deposited in that region had transited through 200 meter depth, while only 0.2 % of the corresponding ^{137}Cs deposition did so (Povinec et al., 1996). In the North Pacific, the Chernobyl-derived radiocesium was detected in the

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sinking particles collected at a depth of 780 m two months after the Chernobyl accident (Kusakabe et al., 1988). The resident time of Pu isotopes in the North Pacific was much shorter than that of radiocesium (Fowler et al., 1983; Honda et al., 2013). Thus Pu isotopes released from the FNDPP accident could be more quickly incorporated into sediments by the scavenging process than Cs, and the determination of Pu isotopes in the sediments should give reliable information about Pu contamination in the marine environment.

To compare the distributions of Pu isotopes in the marine sediments before and after the FNDPP accident, the background dataset needs to be established. Due to the potential applications of Pu isotopes as a chemical tracer for oceanic processes and as a source identifier for radioactive contamination, over the past decades, the distribution of Pu isotopes in the western North Pacific and its marginal seas have been studied intensively (Buesseler, 1997; Dong et al., 2010; Hong et al., 1999; Ito et al., 2007; Kim et al., 2003; Lee et al., 1998, 2003, 2004, 2005; Lindahl et al., 2011; Liu et al., 2011a; Moon et al., 2003; Nagaya and Nakamura, 1992; Oikawa et al., 2011; Otosaka et al., 2006; Pettersson et al., 1999; Wang and Yamada, 2006; Zheng and Yamada, 2005, 2006b,c). For the Japanese near-coastal marine environment, especially the estuaries, leaching of contaminated soils is another potential pathway for radioactive contamination in addition to the atmospheric deposition from the accident plume and the direct release of contaminated plant cooling waters (Bailly du Bois et al., 2012). Before the accident, $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in five estuaries in western and northern Japan had been investigated (Liu et al., 2011b; NIRS, 2010). However, for estuaries in eastern Japan, where it is geographically more possible to be contaminated from the FDNPP accident, the background data for Pu contamination assessment are limited.

In this work, we first measured Pu isotope concentrations in the surface sediments from seven eastern Japan estuaries facing the North Pacific, using samples collected from 2008 to 2010. Then we summarized the published results to establish the comprehensive baseline data of Pu distribution in the western North Pacific and its marginal

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seas. Next we determined the vertical distribution of Pu activities ($^{239+240}$Pu and ^{241}Pu) and Pu atom ratios (^{240}Pu / ^{239}Pu and ^{241}Pu / ^{239}Pu) in five sediment cores, collected in the western North Pacific after the FDNPP accident. Finally, we compared our obtained results with the baseline data to assess the impact of the possible Pu contamination in the marine environment due to the FDNPP accident.

2 Materials and methods

2.1 Study area and sampling

Surface sediment samples (0–2 cm) were collected from seven estuaries (Sagami River, Oyodo River, Yoshino River, Oi River, Naka River, Kitakami River and Kako River) in eastern Japan facing the North Pacific during the years 2008 to 2010. For each estuary, 2–4 samples were collected. Five sediment core samples (ES4, 37°53.00′ N 143°35.00′ E; FS1, 37°20.00′ N 141°25.00′ E; MC5, 37°35.01′ N 141°30.95′ E; MC1, 36°28.97′ N 141°29.93′ E; F1, 36°29.09′ N 141°30.01′ E) were collected by a multiple corer in the western North Pacific Ocean off the FDNPP site during the three cruises of MR-11-05, KH 11-07 and MR-12-02 from July 2011 to July 2012. The core samples were cut into 0.5 cm or 1 cm segments and stored in an on-board refrigerator until brought back to the land-based laboratory. The locations of the surface sediment sampling sites and the sediment core sampling sites in this study are shown in Figs. 1 and 2, respectively.

2.2 Analytical procedure

We modified the method for sample preparation from the literature work (Liao et al., 2008). Briefly, the sediment samples were dried at 105 °C for 24 h. Then they were placed in ceramic crucibles and ashed in a muffle furnace at 600 °C for 5 h. The organic matter content in each sample was determined by the decrease in sample weight

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before and after the ashing process. About 2.0 g dried sample was weighed out and spiked with 100 µL (ca. 1 pg ^{242}Pu) standard ^{242}Pu solution with a traceability to CRM 130 (plutonium spike assay and isotopic standard, New Brunswick Laboratory, USA) as yield monitor. The extraction of Pu was performed in a tightly covered Teflon tube with 20 mL concentrated HNO_3 on a hot plate at 180–200 °C for at least 4 h. A two-stage anion-exchange chromatographic method using AG 1X8 and AG MP-1M resins was employed to separate U and Pu and further purify Pu (Liao et al., 2008). The final sample was prepared in 0.8 mL 4 % HNO_3 media for Pu isotope analysis. The chemical yield of Pu for this employed sample preparation method was estimated to be in the range of 53 % to 85 % with a mean of 66 % ± 10 %.

The measurement of the concentration of Pu isotopes and Pu atomic ratios was done with a double-focusing SF-ICP-MS (Element 2, Thermo Finnigan, Bremen, Germany). An APEX-Q high-efficiency sample introduction system (Elemental Scientific Inc., Omaha, NE, USA) with membrane desolvation unit (ACM) and a conical concentric nebulizer was used. We used the SF-ICP-MS in the low resolution (LR) mode to utilize the maximum instrument sensitivity. In addition, we replaced the normal skimmer cone with a high-efficiency cone (X-cone, Thermo Finnigan) to further increase the sensitivity of the SF-ICP-MS. All the measurements were made in the self-aspirating mode to reduce the risk of contamination by the peristaltic pump tubing. The detailed operation conditions and measurement parameters for this measuring method were described elsewhere (Zheng and Yamada, 2006a). A Pu isotope standard solution (NBS-947) with a known ^{240}Pu / ^{239}Pu atom ratio was used for mass bias correction. Two ocean sediment reference materials (NIST-4357 and IAEA-368) were used for analytical method validation. The analytical results of the reference materials were in good agreement with the certified activity values and the reported Pu atom ratio values (Table 1S, see Supplement).

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3 Results and discussion

3.1 Baseline data for the Pu distribution in the western North Pacific and its marginal seas before the FDNPP accident

Pu isotopes in the western North Pacific and its marginal seas could be attributed to the global fallout and the Pacific Proving Ground (PPG) close-in fallout before the FNDPP accident. The PPG was the name used to describe a number of sites in the Marshall Islands and a few other sites in the Pacific Ocean, used by the United States to conduct nuclear testing between 1946 and 1962. The global fallout Pu has a $^{240}Pu/^{239}Pu$ atom ratio of 0.18 (Bowen et al., 1980; Kelly et al., 1999; Livingston and Povinec, 2002), while the PPG close-in fallout has been characterized by a higher $^{240}Pu/^{239}Pu$ atom ratio (>0.30) (Buesseler, 1999; Diamond et al., 1960; Muramatsu et al., 2001).

The $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in the surface sediments in Japanese estuaries as well as Tokyo Bay, Sagami Bay and Hiroshima Bay are summarized in Table 1 and the $^{239+240}Pu$ activities are presented in Fig. 1. As shown in Fig. 1, the concentration of $^{239+240}Pu$ activity in the Japanese estuaries is relatively low, ranging from 0.003 to 1.191 $mBq\,g^{-1}$. In particular, for the estuaries off the eastern coast of Japan facing the North Pacific, investigated in this study, the concentration of $^{239+240}Pu$ activities were typically below 0.400 $mBq\,g^{-1}$ (Table 2S, see Supplement). The highest concentration (5.81 $mBq\,g^{-1}$) of $^{239+240}Pu$ activity was reported in the surface sediment of Sagami Nada (Zheng and Yamada, 2004). For all the estuary sediments investigated in this study, the ^{241}Pu activity was below the detection limit (2 $mBq\,g^{-1}$) of the SF-ICP-MS.

The $^{240}Pu/^{239}Pu$ atom ratios in the Japanese near-coastal surface sediments are shown in Fig. 3, and they ranged from 0.170 to 0.270. All the values were generally higher than the value of global fallout, except for CM-04 and CM-06-08 in northern Japan. Zheng and Yamada (2006b) suggested CM-04 and CM-06-08 received only trace amounts of PPG close-in fallout Pu via atmospheric deposition. An average $^{240}Pu/^{239}Pu$ atom ratio of 0.231 ± 0.025 ($n = 36$) was obtained for the Japanese

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eastern estuaries facing the North Pacific, which indicated the mixing of global fallout Pu and PPG close-in fallout Pu.

A more comprehensive summarization about the distribution of Pu isotopes in the western North Pacific and its marginal seas is given in Table 2. The concentration of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in the surface sediments ranged from 0.002 to 5.38 mBq g^{-1} and from 0.150 to 0.281, respectively, except in the surface sediment collected near the Bikini Atoll, where the high concentration of 12.5 mBq g^{-1} and high atom ratio of 0.336 have been reported (Buesseler, 1997; Lee et al., 2005). In the Okinawa Trough and Sagami Bay, the ^{240}Pu/^{239}Pu atom ratios were found to increase with depth with a maximum of 0.30–0.33 in the deeper layer (> 5 cm) sediments (Lee et al., 2004; Wang and Yamada, 2005; Zheng and Yamada, 2006b), indicating clearly the early deposition of the PPG close-in fallout Pu.

Historically, ^{241}Pu was released into the environment by the nuclear weapon testing mainly conducted 50 yr ago and its half-live is relatively short (14.4 yr), so the current activity of ^{241}Pu in the environment is quite low. Yamamoto et al. (1990) determined the Pu isotopes in the sediments collected from the Nyu Bay in the Japan Sea, and they found that the ^{241}Pu activities and the ^{241}Pu/$^{239+240}$Pu activity ratios ranged from 4.5 mBq g^{-1} to 7.5 mBq g^{-1} and from 1.2 to 1.9, respectively. (For all the values related to ^{241}Pu discussed here, decay corrections have been made to 11 March 2011.) Similarly, Zheng and Yamada (2008) observed a ^{241}Pu activity of 8.4 mBq g^{-1} and a ^{241}Pu/$^{239+240}$Pu activity ratio of 1.2 at a depth of 18–20 cm in a sediment core collected from Sagami Bay. More comprehensive investigations were conducted by the Japanese government on the distribution of Pu isotopes in the surface marine sediments off Japan, and the results for the ^{241}Pu activity and ^{241}Pu/$^{239+240}$Pu activity ratio in the surface sediments were typically below 3.3 mBq g^{-1} and 1.1, respectively (MEXT, 2008). Due to the influence of the PPG close-in fallout, high ^{241}Pu activities (up to 19.3–33.4 mBq g^{-1}) and the ^{241}Pu/$^{239+240}$Pu activity ratios (2.2–2.7) were observed in the sediments near the Bikini Atoll (Lee et al., 2005).

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For the inventory of $^{239+240}$Pu, a wide range of values (1.38–693.33 Bq m^{-2}) have been observed. The vertical distribution of Pu isotopes in the sediments varied significantly due to the difference of bottom topography and sedimentation dynamics. For example, the enhanced particle scavenging in the continental margin resulted in higher Pu inventories in the sediments in the Okhotsk Sea (81–271 Bq m^{-2}) compared to those from the global fallout areas (Zheng and Yamada, 2006b). However, for the western North Pacific, the inventory of $^{239+240}$Pu ranged from 2.8 to 71.8 Bq m^{-2} (Moon et al., 2003). Thus the inventory of $^{239+240}$Pu in the sediments cannot provide a direct index for source identification. Isotopic composition information should be combined with the inventory values to reach an accurate conclusion.

In summary, the $^{239+240}$Pu activity and the ^{240}Pu / ^{239}Pu atom ratio in the upper layer sediments (< 5 cm) in the western North Pacific and its marginal seas off Japan before the FDNPP accident could be considered as lower than 5.81 mBq g^{-1} and 0.28, respectively. For the deeper layer sediments (> 5 cm), the ^{240}Pu / ^{239}Pu atom ratio could reach about 0.30 due to the earlier deposition of the PPG close-in fallout. The background values for ^{241}Pu activity and ^{241}Pu / $^{239+240}$Pu activity ratio in the sediments in the Japanese coastal areas could be lower than 10 mBq g^{-1} and 3, respectively. The inventories of Pu vary significantly depending on the different bottom topography and sedimentation dynamics.

3.2 Vertical distribution of Pu in sediment cores from the western North Pacific after the FDNPP accident

3.2.1 Vertical distribution of Pu activities and Pu inventories

The analytical results of Pu isotopes in the sediment core samples are summarized in Table 3 and plotted in Fig. 4. The surface $^{239+240}$Pu activities in the sediments of ES4, MC1, MC5 and FS1 ranged from 0.476 to 2.809 mBq g^{-1}. When we compare them with the baseline data of the adjacent open oceans of the Pacific Ocean, there was no significant increase after the accident (Zheng et al., 2012b). For the F1 station, the surface

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$^{239+240}$Pu activity was 1.774 mBq g^{-1}, which was comparable to that (1.580 mBq g^{-1}) of the MC1 station. The F1 station is close to the MC1 station and the F1 sampling date (7 July 2012) was one year after the MC1 sampling date (18 July 2011). The similar surface $^{239+240}$Pu activity for these two stations indicated that there was no remarkable change for $^{239+240}$Pu activity in the surface sediments in the western North Pacific about 110 km southeast of the FDNPP during the last year (July 2011 to July 2012). The highest $^{239+240}$Pu activity (3.079 mBq g^{-1}) was observed in the upper layer (1–2 cm) of FS1, which was lower than the upper limit of the baseline data (5.38 mBq g^{-1}) before the FNDPP accident as we concluded in Sect. 3.1.

As shown in Fig. 4, the vertical profiles of $^{239+240}$Pu activities in the five sediment cores showed different patterns. For the cores of MC1 and FS1, a surface $^{239+240}$Pu maximum was observed and the $^{239+240}$Pu activities decreased with core depth. For the cores of F1 and MC5, approximately uniform $^{239+240}$Pu activities were found from the surface to a depth of 5 cm and 8 cm, respectively. Similar observations were reported by Zheng et al. (2006b) for the $^{239+240}$Pu distribution in two sediment cores (CM-03 and CM-04) from the western North Pacific (Fig. 4). Sediment particles mixing by bioturbation and/or physical factors after the deposition are thought to be the controllers of the surface mixing layer (SML) (Zheng et al., 2006b). For the marine environment near the Japanese coast, the M9.0 East Japan Earthquake was suggested to be another important factor that caused the displacement of the seafloor and resulted in turbulent diffusion of the seafloor sediments (Kawagucci et al., 2012). The SML depth (8 cm) in CM-04 was comparable with that of F1 and MC5. A subsurface maximum of $^{239+240}$Pu activity was found in the sediment core of ES4. Usually, in areas with a high sedimentation rate, the subsurface maximum has corresponded to the year 1963, when the largest global fallout of Pu occurred (Hong et al., 1999). Moon et al. (2003) reported relatively low sedimentation rates (0.12–0.35 cm kyr^{-1}) in the western North Pacific after they investigated the ^{14}C and ^{230}Th$_{ex}$ distributions in the sediment profiles. As the water depth exceeds 5000 m at the ES4 station, the sedimentation rate is not a key factor controlling the vertical distribution of Pu isotopes there. However, the biological

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activity in the sea floor in the western North Pacific is high, evidencing a high bioturbation rate in the surficial marine sediment (Harada and Shibamoto, 2002). The penetration depth of radionuclides into the seabed can be measured and then related to mixing rate and sedimentation rate by incorporating the dispersion-advection equation (Demaster et al., 1985):

$$Z = \sqrt{2D_b T} + ST \quad (1)$$

where Z is the penetration depth (cm), D_b is the mixing coefficient ($cm^2\,yr^{-1}$), T is the elapsed time (yr) since deposition and S is the sedimentation rate ($cm\,yr^{-1}$). The particle mixing coefficient ranges from 0.02 to 1.00 $cm^2\,yr^{-1}$ in the western North Pacific (Moon et al., 2003). If we assume that 48 yr had passed since 1963, the penetration depth of Pu in the western North Pacific can be roughly estimated to be 1.4–9.9 cm by Eq. (1). This is consistent with the result (8 cm) we observed at the ES4 station. Similarly, Harada and Shibamoto (2002) observed that the penetration depth of excess ^{210}Pb was 7.2 cm in the same sea area of ES4.

High ^{241}Pu activities (4.5–34.8 $mBq\,g^{-1}$) and ^{241}Pu / $^{239+240}$Pu activity ratio (107.8) in the surface soil and litter samples from the 20–30 km zone around the FNDPP were observed, evidencing the contamination from the accident (Zheng et al., 2012b). However, for the five sediment cores investigated in this study, a detectable ^{241}Pu activity (7.42 $mBq\,g^{-1}$) only occurred at the ES4 station at a depth of 3–4 cm where the $^{239+240}$Pu activity peaked. This value was typically in the range (< 10 $mBq\,g^{-1}$) of the background database before the FNDPP accident. Meanwhile, the ^{241}Pu / $^{239+240}$Pu activity ratio (2.5), comparable with reported results before the accident, was significantly lower than that derived from the FDNPP accident.

Organic matter plays an important role regarding the adsorption of radionuclides in the sediments. A study about the distribution of ^{137}Cs in the marine sediments off the Ibaraki coast following the FNDPP accident showed that ca. 20 % of the ^{137}Cs was organically bounded (Otosaka and Kawamura, 2013). It was reported that about 40 %–80 % of the plutonium isotopes could be organic matter-bound in the marine sediments

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(Outola et al., 2009; Qiao et al., 2012). The vertical profiles of $^{239+240}$Pu activity and organic matter content in the sediment core samples are shown in Fig. 5. For the cores of ES4, MC1, MC5 and F1, the organic matter content stayed almost uniform from the surface to the deeper layer, while for the core of FS1, the organic matter content was higher in the surface layer (< 5 cm) than that in the deeper layer (> 5 cm). The high concentration of $^{239+240}$Pu activity in the surface layer sediments of FS1 could be partly due to the relatively high concentration of organic matter. However, no significant relationship between the vertical distribution of $^{239+240}$Pu activity and the distribution of organic matter content was observed for the other four sediment cores.

Plutonium inventory in sediment columns reflects the source function, transport of Pu in water and on particles, and the mixing process; thus it is an important parameter for geochemical study of Pu. The inventory of Pu in a sediment core can be estimated by summing the Pu concentration in each layer of the sediment (Livingston et al., 2001):

$$I_s = \sum_{i=1}^{M} S_i D_i (U_i - L_i) \quad (2)$$

where I_s is the Pu inventory in a 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^{-1}, dry weight), D_i is the dry bulk density of layer i (kg m^{-3}), U_i is the upper boundary of layer i (m) and L_i is the lower boundary of layer i (m).

The obtained Pu inventories for ES4, FS1, MC5, MC1 and F1 were 29.3, 131.7, 95.5, 32.6 and 77.2 Bq m^{-2}, respectively. As shown in Fig. 6, the inventories varied significantly. The lowest inventory was observed at ES4 station and the relatively low Pu accumulation may be due to the deep water depth and the low sedimentation rate. The highest inventory was found at the FS1 station, which was the nearest one to the Fukushima coast among the five sampling stations. High Pu inventories seem to be a common phenomenon in the coastal sediments in the western North Pacific and its

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marginal seas. For example, Lee et al. (2004) observed extremely high Pu inventories (201–693 $Bq\,m^{-2}$) in the Southern Okinawa Trough. Zheng et al. (2005, 2006a) reported that the Pu inventories in the Japan Sea and the Okhotsk Sea ranged from 5.7 to 241 $Bq\,m^{-2}$ and from 81 to 271 $Bq\,m^{-2}$, respectively. The advective westward transport of Pu from the Marshall Islands by the North Equatorial Current followed by northward transport of Kuroshio Current was suggested to be an important factor causing the high Pu input (Lee et al., 2004; Zheng et al., 2004, 2006c). The Pu inventory (77.2 $Bq\,m^{-2}$) at the F1 station is more than two times that (32.6 $Bq\,m^{-2}$) at the MC1 station. These two stations are off the Ibaraki coast and are very close geographically. In the coastal area of Ibaraki, Otosaka and Kawamura (2013) investigated the distribution of radiocesium in the sediments collected from near-coast to off-coast sites over some time after the FDNPP accident; and they found that the radiocesium, first appeared in the near-coast region, and then was transported to the off-coast region through suspended fine particles in a high-turbidity layer. However, the Pu inventory variation between the F1 and MC1 stations could not be simply explained as Pu accumulation from the near-coast region during the last year (July 2011 to July 2012). A more comprehensive comparison can be done with the results of the CM-03 station. The CM-03 station, which is located 200 km southeast of the FDNPP, is the only area close to the FNDPP for which the vertical distribution of $^{239+240}Pu$ activity before the accident is known. In the CM-03 sediment core, the Pu inventory was reported to be 74 $Bq\,m^{-2}$ (Zheng and Yamada, 2006b), which was comparable with the result of F1.

3.2.2 Vertical distribution of Pu atom ratios and the possible sources of Pu

Pu atom ratios ($^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$) have been demonstrated to be good indicators for the Pu source identification. For instance, the atom ratio of $^{240}Pu/^{239}Pu$ in weapon grade Pu is typically around 0.05 and higher ratios can be expected with higher neutron fluxes associated with an increase in the yield of a nuclear detonation (Buesseler, 1997; Koide et al., 1985). For a nuclear reactor, the Pu atom ratios can vary from 0.23 to 0.65 depending on the fuel characteristics, type and design of the reactor,

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operational conditions of the plant and cooling time since the fuel was last irradiated in the reactor (Ketterer and Szechenyi, 2008; Schwantes et al., 2012; Taylor et al., 2001). The $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ atom ratio fingerprints for the Pu isotopes released from the FDNPP accident were suggested to be 0.303–0.330 and 0.103–0.135, respectively, and they were significantly higher than the global fallout values (Zheng et al., 2012b).

As shown in Table 3 and Fig. 4, the $^{240}Pu/^{239}Pu$ atom ratios in the five core samples have four major characteristics: (1) the ratios ranged from 0.188 to 0.293 and were typically higher than the reported global fallout value (0.18); (2) all the cores showed a nearly uniform distribution of $^{240}Pu/^{239}Pu$ atom ratio in the upper sediment layers; (3) the $^{240}Pu/^{239}Pu$ atom ratios increased from a definite depth in the cores of MC1, F1 and FS1, while the ratios stayed almost uniform in the cores of MC5 and ES4; and (4) the inventory-weighted $^{240}Pu/^{239}Pu$ atom ratio (0.202) at the ES4 station was lower than the ratios of the other four stations (FS1, 0.236; MC5, 0.240; MC1, 0.240; F1, 0.243).

We can see that almost all the sediment samples investigated in this study showed higher $^{240}Pu/^{239}Pu$ atom ratio than the global fallout. However, higher $^{240}Pu/^{239}Pu$ atom ratio was observed in a wide range of sea areas in the western North Pacific before the FNDPP accident due to the presence of the PPG close-in fallout as discussed in Sect. 3.1. As the majority of the tests at the PPG were conducted at or just above the earth's surface and the maximum deposition occurred in 1954, the injection of Pu from the PPG as tropospheric deposition to the western North Pacific happened before the global fallout injection. Therefore, the higher $^{240}Pu/^{239}Pu$ atom ratios in the deeper layers in the sediment cores of MC1, F1 and FS1 retained their validity as a record for the PPG close-in fallout. In Sagami Bay, on the western North Pacific margin, a similar distribution for the $^{240}Pu/^{239}Pu$ atom ratio in the sediment core was also observed (Zheng and Yamada, 2004); that is, a nearly uniform $^{240}Pu/^{239}Pu$ atom ratio of about 0.24 distributed from the surface down to the layer 12–14 cm deep, and a ratio higher than 0.27 starting from the layer at 16–18 cm depth. Our observed results provided

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further evidence for the hypothesize that the oceanic processes (North Equatorial Current and the Kuroshio Current) transported the PPG close-in fallout Pu westwards as far as the Japanese coast. The uniform distribution of the $^{240}Pu/^{239}Pu$ atom ratio in the sediment cores of MC5 and ES4 could be due to the mixing process caused by bioturbation and/or physical factors. Unlike the other four stations, the ES4 station is located in the pathway of the southward flowing Oyashio Current, which carries water bearing global fallout Pu. The CM-04 station is located in northern Japan in the western North Pacific and is affected by the Oyashio Current as well, and it showed a typical distribution of $^{240}Pu/^{239}Pu$ atom ratios which was the same as the global fallout. Thus the trace of PPG-derived Pu at the ES4 station could be from direct fallout in that region when the nuclear weapon tests were conducted.

We have concluded in Sect. 3.1 that the $^{240}Pu/^{239}Pu$ atom ratio in the western North Pacific was typically below 0.28 in the upper layer (< 5 cm) sediments before the FDNPP accident. For all the sediment cores investigated in this study, the upper layer sediment $^{240}Pu/^{239}Pu$ atom ratio was below the background baseline limit. The deeper layer sediments, characterized by high $^{240}Pu/^{239}Pu$ atom ratio in FS1, MC1 and F1 were due to the early deposition of the PPG close-in fallout. Thus no significant amounts of Pu injection from the FDNPP accident, which has a high $^{240}Pu/^{239}Pu$ atom ratio (> 0.30), were observed in the investigated regions. The data on the distribution of ^{241}Pu give further support to this conclusion. No ^{241}Pu was detected for all the samples except one at the ES4 station with a concentration of 7.42 mBq g^{-1} and a $^{241}Pu/^{239}Pu$ atom ratio of 0.0029. This $^{241}Pu/^{239}Pu$ atom ratio was similar to the result (0.0033) we observed in the sediment at ES2 (Zheng et al., 2012a) and results (0.0031–0.033) in the sediment near the Bikini Atoll reported by Lee et al. (2005). As the Fukushima-derived $^{241}Pu/^{239}Pu$ atom ratio was much higher than the observed result, the presence of ^{241}Pu in ES4 further evidenced the PPG source contribution to that region.

Presuming that the $^{240}Pu/^{239}Pu$ atom ratio for global fallout is 0.18 (Kelly et al., 1999), and that the representative $^{240}Pu/^{239}Pu$ atom ratio for the PPG close-in fallout

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is 0.36 (Buesseler, 1999; Diamond et al., 1960; Muramatsu et al., 2001), we could estimate the contribution of Pu source to the Pu inventory in the western North Pacific by using a two-end member-mixing model (Krey et al., 1976):

$$Y = \frac{(\mathrm{Pu})_\mathrm{P}}{(\mathrm{Pu})_\mathrm{G}} = \frac{(R_\mathrm{G} - R_\mathrm{S})(1 + 3.66R_\mathrm{P})}{(R_\mathrm{S} - R_B)(1 + 3.66R_\mathrm{G})} \quad (3)$$

where (Pu) is the $^{239+240}$Pu activity; R is the ^{240}Pu / ^{239}Pu atom ratio; and the subscript P, G and S refer to PPG close-in fallout, global fallout and measured sediment samples, respectively. Via Eq. (2), we calculated the inventory-weighted percentages of the PPG close-in fallout in each sediment core.

As shown in Fig. 7, we found that the PPG close-in fallout contributed a nearly constant percentage (ca. 40 %) of the Pu inventory in the sediment cores in the western North Pacific off the eastern coast of Japan except for ES4. As discussed above, ES4 is located in the pathway of the Oyashio Current, and received only a trace of directly deposited Pu from the PPG close-in fallout. For stations F1 and MC1, although the Pu inventories in these two close stations showed significant variation, the sources of Pu in them were identical (the PPG close-in fallout Pu and global fallout Pu), suggesting no detectable Pu contamination originating from the FDNPP accident injected to that region during the year since the accident.

4 Conclusions

The vertical profiles of Pu isotopes in five sediments cores obtained in the western North Pacific from July 2011 to July 2012 after the FNDPP accident as well as surface sediments in seven Japanese estuaries collected before the FNDPP accident were investigated. We first established the comprehensive background baseline data for the Pu distribution in the sediments in the western North Pacific and its marginal seas before the FNDPP accident. Then we compared the results of the five sediment cores with the baseline data. We could not identify any extra Pu injection from the FDNPP

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accident in the marine sediments collected outside the 30 km zone from the plant site up to the time of sampling. The global fallout and the PPG close-in fallout Pu were the two main sources for the Pu contamination in the marine environment outside the 30 km zone. However, as Pu isotopes are particle-reactive, the Pu contamination situation in the marine environment within the 30 km zone around the FDNPP needs further investigation before a more comprehensive conclusion can be reached.

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/10/643/2013/bgd-10-643-2013-supplement.pdf.

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Table 1. Summary of the distributions of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in the surface sediments (0–2 cm) from Japanese estuaries.

Area	Location	Sampling date	Surface $^{239+240}$Pu activity (mBq g^{-1})	Surface ^{240}Pu/^{239}Pu atom ratio	Reference
Mabechi River Estuary	40°32–35′ N, 141°31–36′ E	2007	0.11–0.811	0.204–0.249	NIRS (2010)
Mogami River Estuary	38°55–57′ N, 139°46–47′ E	2007	0.187–0.478	0.201–0.233	NIRS (2010)
Yura River Estuary	35°31–35′ N, 135°16–20′ E	2007	0.122–1.191	0.216–0.248	NIRS (2010)
Kuma River Estuary	32°22–27′ N, 130°28–31′ E	2007	0.537–0.708	0.228–0.242	NIRS (2010)
Tokyo Bay	35°12–19′ N, 139–140° E	1988	1.05–2.66	ND	Yamada and Nagaya (2000)
Sagami Bay	35°12–35′ N, 139–140° E	1990–1991	0.77–5.81	0.232–0.244	Zheng and Yamada (2004)
Hiroshima Bay	34°11.5–18.5′ N, 132°20.5–22.0′ E	2009	0.556–0.677	0.224–0.239	Liu et al. (2011b)
Sagami River Estuary	35°18–20′ N, 139°22–23′ E	2008	0.010–0.077	0.258–0.261	This study
Oyodo River Estuary	31°53–54′ N, 131°28–30′ E	2009	0.196–0.346	0.254–0.270	This study
Yoshino River Estuary	34°4–5′ N, 134°36–38′ E	2009	0.060–0.114	0.209–0.231	This study
Oi River Estuary	34°46′ N, 138°18–19′ E	2009	0.003–0.005	0.236–0.264	This study
Naka River Estuary	36°20–21′ N, 140°35–38′ E	2009	0.117–0.351	0.243–0.246	This study
Kitakami River Estuary	38°34–35′ N, 141°28–30′ E	2010	0.050–0.172	0.205–0.214	This study
Kako River Estuary	34°42–44′ N, 134°47–49′ E	2010	0.090–0.129	0.202–0.220	This study

ND: Not determined.

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Table 2. Summary of the distributions of $^{239+240}$Pu activity and ^{240}Pu / ^{239}Pu atom ratio in sediments from the western North Pacific and its marginal seas.

Area	Location	Sampling date	Water depth (m)	Surface $^{239+240}$Pu activity (mBq g^{-1})[a]	Surface ^{240}Pu / ^{239}Pu atom ratio[a]	Vertical ^{240}Pu / ^{239}Pu atom ratio range	$^{239+240}$Pu inventory (Bq m^{-2})	Reference
West Caroline Basin	2–5° N, 137° E	1992	4157–4629	0.08–0.2	ND	ND	6.82–8.95	Moon et al. (2003)
Solu Sea	8.9° N, 121.5° E	1996	4988	0.015–0.508	0.257–0.281	0.257–0.281	1.38	Dong et al. (2010)
Near Bikini Atoll	11°26′ N, 164°52′ E	1997	ND	9.0–12.5	0.239–0.242	0.133–0.388	130	Lee et al. (2005)
Near Bikini Atoll	7°03.2′ N, 164°47.3′ E	1978	5925	ND	0.336 ± 0.012	ND	ND	Buesseler (1997)
South China Sea	15.5° N, 115.3° E	1997	4234	0.099–0.157	0.228–0.243	0.227–0.300	3.75	Dong et al. (2010)
Northwest-central Basin	15–35° N, 145–159° E	1997	5390–5924	0.15–5.38	ND	ND	2.8–71.8	Moon et al. (2003)
Southern Okinawa Trough	24–25° N, 122–123° E	2000–2003	> 1000	1.3–3.0	0.23–0.25	0.2–0.33	201.67–693.33	Lee et al. (2004)
Okinawa Trough	27–29° N, 126–128° E	1992–1995	999–1080	0.775–2.496	0.210–0.261	0.210–0.320	32.5–47.2	Wang and Yamada (2005)
East China Sea	28–32° N, 123–127° E	1992–1995	50–127	0.250–1.160	0.251–0.261	0.236–0.297	60.9–101	Wang and Yamada (2005)
Yangtze River Estuary	29–33° N, 121–125° E	2006	10–70	0.05–0.76	0.22–0.26	0.190–0.319	387	Liu et al. (2011a)
East China Sea	26–30° N, 127–131° E	1993–1994	1830–1870	1.25–3.56	0.218–0.235	0.218–0.247	14.8–42	Zheng and Yamada (2006c)
East China Sea and the Yellow Sea	25–35° N, 122–124° E	1987	37–2170	0.107–0.467	ND	ND	8.9–79.9	Nagaya and Nakamura (1992)
East Sea	20–45° N, 140–145° E	1995	1170–2600	2.00–3.73	ND	ND	59.4–65.5	Lee et al. (1998)
Japan Sea/East Sea	37–41° N, 129–134° E	1993–1997	1002–3400	0.98–2.74	ND	ND	13–68	Hong et al. (1999)
Mikata Five Lakes and Nyu Bay, Japan Sea	35–36° N, 135–136° E	1986–1988	11–40	1.36–5.71	ND	ND	109–347	Yamamoto et al. (1990)
Japan Sea	37–46° N, 135–140° E	1998	91–3670	0.07–2.65	0.160–0.223	0.139–0.241	5.7–241	Zheng and Yamada (2005)
Japan Sea	35–43° N, 130–135° E	1994–1995	1512–3570	0.011–1.79	ND	ND	ND	Pettersson et al. (1999)
Japan Sea	36–44° N, 131–139° E	1997–2000	359–3600	ND	ND	ND	0.5–67.7	Ito et al. (2007)
Japan Sea	36–42° N, 131–133° E	1998–2002	359–3560	0.002–1.9	ND	ND	4.7–37	Otosaka et al. (2006)
Coast of the Korean Peninsula	33–40° N, 125–130° E	2000	ND	0.02–1.72	0.15–0.23	0.15–0.23	ND	Kim et al. (2003)
Okhotsk Sea	44–46° N, 145–146° E	1998	1214–3053	2.10–3.61	0.170–0.204	0.165–0.215	81–271	Zheng and Yamada (2005)
Okhotsk Sea	51°00′ N, 151°00′ E	1995	1300	0.92–1.12	ND	ND	40	Lee et al. (2003)
Okhotsk Sea	47–55° N, 140–151° E	1995	136–1340	0.30–0.77	ND	ND	ND	Pettersson et al. (1999)
Off Japanese coast in the Pacific	35–43° N, 141–146° E	1998	2323–3318	0.56–3.80	0.140–0.242	0.152–0.270	62–74	Zheng and Yamada (2006b)
Aomori Sea	40–42° N, 141–142° E	1991–2005	50–1100	0.485–4.00	0.218–0.248	ND	ND	Oikawa et al. (2011)
NW Pacific	33–40° N, 155–163° E	1995	2000–5500	0.26–1.99	ND	ND	ND	Pettersson et al. (1999)

[a] surface sediment 0–2 cm.
ND: Not determined.

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Table 3. Results of Pu activities and Pu atom ratios in the sediment cores.

Sample interval (cm)	Organic matter content (%)	$^{239+240}$Pu activity (mBq g^{-1})	^{241}Pu activity (mBq g^{-1})	^{240}Pu / ^{239}Pu atom ratio	^{241}Pu / ^{239}Pu atom ratio	$^{239+240}$Pu inventory (Bq m^{-2})
KH 11-07 ES4 (37°53.00′ N, 143°35.00′ E, 18 Jul 2011, 5400 m water depth, 22 cm core length)						
0–1[a]	17.9	1.231 ± 0.026	ND	0.188 ± 0.009	ND	29.3
1–2	13.9	2.059 ± 0.039	ND	0.210 ± 0.006	ND	(0–8 cm)
2–3	13.1	2.795 ± 0.052	ND	0.203 ± 0.006	ND	
3–4	14.8	2.923 ± 0.063	7.42 ± 0.61[b]	0.196 ± 0.005	0.0027 ± 0.0002[b]	
4–5	12.4	1.025 ± 0.042	ND	0.212 ± 0.004	ND	
5–6	11.6	0.050 ± 0.004	ND	0.198 ± 0.025	ND	
6–7	11.1	0.007 ± 0.002	ND	ND	ND	
7–8	10.9	0.002 ± 0.001	ND	ND	ND	
KH 11-07 FS1 (37°20.00′ N, 141°25.00′ E, 2 Aug 2011, 150 m water depth, 24 cm core length)						
0–1[a]	15.3	2.809 ± 0.040	ND	0.224 ± 0.006	ND	131.7
1–2	12.1	3.079 ± 0.238	ND	0.228 ± 0.036	ND	(0–20 cm)
2–3	10.1	2.494 ± 0.261	ND	0.233 ± 0.011	ND	
3–4	8.7	2.144 ± 0.134	ND	0.237 ± 0.014	ND	
4–5	8.0	1.970 ± 0.109	ND	0.232 ± 0.023	ND	
5–6	7.7	1.623 ± 0.105	ND	0.240 ± 0.013	ND	
6–7	8.2	1.663 ± 0.053	ND	0.228 ± 0.010	ND	
7–8	8.9	1.572 ± 0.053	ND	0.232 ± 0.011	ND	
8–9	9.0	1.361 ± 0.050	ND	0.236 ± 0.012	ND	
9–10	7.9	0.674 ± 0.053	ND	0.240 ± 0.043	ND	
10–11	8.1	0.466 ± 0.032	ND	0.243 ± 0.028	ND	
11–12	8.2	0.354 ± 0.030	ND	0.258 ± 0.039	ND	
12–13	8.4	0.473 ± 0.030	ND	0.275 ± 0.037	ND	
13–14	8.0	0.487 ± 0.028	ND	0.286 ± 0.030	ND	
14–15	7.7	0.042 ± 0.005	ND	0.262 ± 0.051	ND	
15–16	8.0	0.027 ± 0.001	ND	ND	ND	
16–17	8.1	0.009 ± 0.002	ND	ND	ND	
17–18	8.5	0.008 ± 0.002	ND	ND	ND	
18–19	8.2	0.007 ± 0.001	ND	ND	ND	
19–20	8.3	0.005 ± 0.002	ND	ND	ND	

[a] Data for 0–1 cm are cited from Zheng et al. (2012b).
[b] ^{241}Pu decay corrected to 15 October 2012.
ND: Not detected.

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Table 3. Continued.

Sample interval (cm)	Organic matter content (%)	$^{239+240}$Pu activity (mBq g^{-1})	^{241}Pu activity (mBq g^{-1})	^{240}Pu / ^{239}Pu atom ratio	^{241}Pu / ^{239}Pu atom ratio	$^{239+240}$Pu inventory (Bq m^{-2})
MR 11-05 MC5 (37°35.01′ N, 141°30.95′ E, 19 Jul 2011, 141 m water depth, 19 cm core length)						
0–1[a]	3.9	0.476 ± 0.008	ND	0.255 ± 0.009	ND	95.5
1–2	4.1	0.486 ± 0.015	ND	0.243 ± 0.015	ND	(0–19 cm)
2–3	4.4	0.483 ± 0.011	ND	0.242 ± 0.011	ND	
3–4	4.5	0.504 ± 0.014	ND	0.242 ± 0.014	ND	
4–5	4.4	0.488 ± 0.013	ND	0.248 ± 0.012	ND	
5–6	4.1	0.469 ± 0.016	ND	0.244 ± 0.015	ND	
6–7	3.9	0.450 ± 0.013	ND	0.243 ± 0.008	ND	
7–8	4.7	0.459 ± 0.011	ND	0.237 ± 0.013	ND	
8–9	3.5	0.327 ± 0.010	ND	0.247 ± 0.018	ND	
9–10	3.6	0.286 ± 0.008	ND	0.241 ± 0.014	ND	
10–11	3.0	0.216 ± 0.009	ND	0.250 ± 0.017	ND	
11–12	3.1	0.135 ± 0.005	ND	0.247 ± 0.016	ND	
12–13	3.5	0.075 ± 0.004	ND	0.241 ± 0.028	ND	
13–14	4.2	0.092 ± 0.004	ND	0.236 ± 0.026	ND	
14–15	4.1	0.157 ± 0.006	ND	0.214 ± 0.013	ND	
15–16	4.1	0.216 ± 0.009	ND	0.201 ± 0.017	ND	
16–17	4.2	0.251 ± 0.007	ND	0.231 ± 0.011	ND	
17–18	4.3	0.225 ± 0.007	ND	0.237 ± 0.016	ND	
18–19	4.0	0.258 ± 0.010	ND	0.240 ± 0.024	ND	
MR 11-05 MC1 (36°28.97′ N, 141°29.93′ E, 18 Jul 2011, 1327 m water depth, 13 cm core length)						
0–1[a]	7.8	1.580 ± 0.026	ND	0.236 ± 0.007	ND	32.6
1–2	8.4	1.372 ± 0.023	ND	0.241 ± 0.010	ND	(0–9 cm)
2–3	7.6	0.536 ± 0.014	ND	0.243 ± 0.013	ND	
3–4	8.8	0.159 ± 0.018	ND	0.241 ± 0.042	ND	
4–5	9.9	0.045 ± 0.004	ND	0.266 ± 0.051	ND	
5–6	8.5	0.016 ± 0.003	ND	0.293 ± 0.080	ND	
6–7	8.5	0.008 ± 0.002	ND	ND	ND	
7–8	9.6	0.006 ± 0.001	ND	ND	ND	
8–9	9.1	0.005 ± 0.001	ND	ND	ND	

[a] Data for 0–1 cm are cited from Zheng et al. (2012b).
[b] ^{241}Pu decay corrected to 15 October 2012.
ND: Not detected.

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Table 3. Continued.

Sample interval (cm)	Organic matter content (%)	$^{239+240}$Pu activity (mBq g^{-1})	^{241}Pu activity (mBq g^{-1})	^{240}Pu / ^{239}Pu atom ratio	^{241}Pu / ^{239}Pu atom ratio	$^{239+240}$Pu inventory (Bq m^{-2})
MR 12-02 F1 (36°29.09′ N, 141°30.01′ E, 7 Jul 2011, 1322 m water depth, 13.2 cm core length)						
0–0.5	8.7	1.774 ± 0.054	ND	0.240 ± 0.006	ND	77.2
0.5–1	9.5	1.910 ± 0.024	ND	0.238 ± 0.004	ND	(0–13.2 cm)
1–1.5	5.9	1.591 ± 0.026	ND	0.237 ± 0.007	ND	
1.5–2	5.9	1.441 ± 0.032	ND	0.241 ± 0.008	ND	
2–3	7.3	1.492 ± 0.021	ND	0.238 ± 0.005	ND	
3–4	7.6	1.462 ± 0.021	ND	0.235 ± 0.006	ND	
4–5	8.2	1.337 ± 0.018	ND	0.232 ± 0.007	ND	
5–6	7.0	0.774 ± 0.014	ND	0.234 ± 0.009	ND	
6–7	7.3	0.209 ± 0.009	ND	0.274 ± 0.021	ND	
7–8	7.3	0.264 ± 0.013	ND	0.290 ± 0.029	ND	
8–9	9.4	0.588 ± 0.011	ND	0.274 ± 0.010	ND	
9–10	7.5	0.219 ± 0.008	ND	0.284 ± 0.023	ND	
10–12	7.5	0.147 ± 0.011	ND	0.291 ± 0.044	ND	
12–13.2	6.8	0.364 ± 0.012	ND	0.256 ± 0.019	ND	

[a] Data for 0–1 cm are cited from Zheng et al. (2012b).
[b] ^{241}Pu decay corrected to 15 October 2012.
ND: Not detected.

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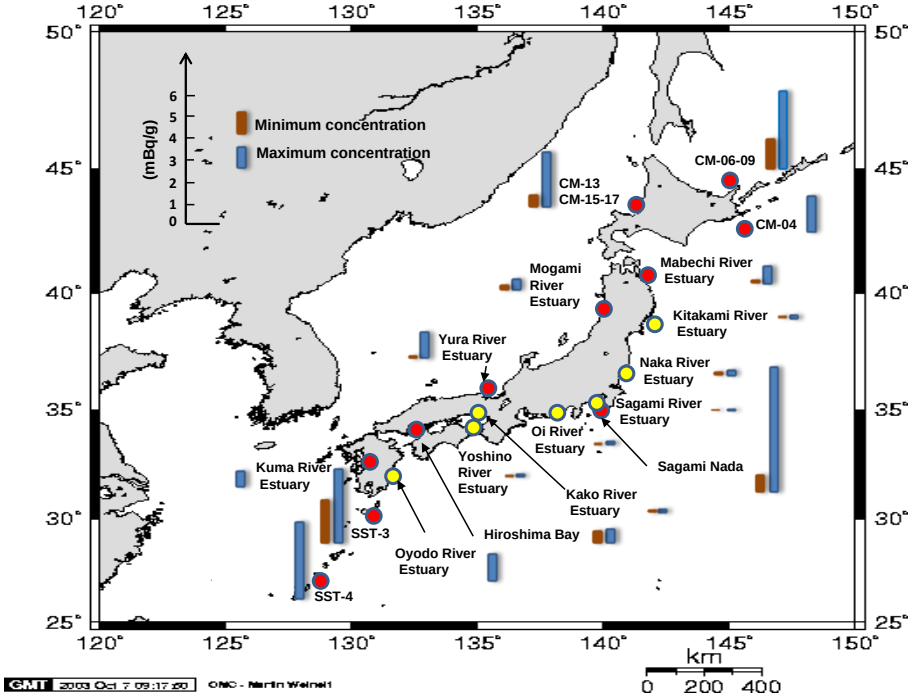

Fig. 1. Map showing the Japanese estuaries and the Pu concentration ($mBq\,g^{-1}$) in the surface sediments (0–2 cm) in the Japanese near-coast marine environment. The yellow circled sites represent the sampling sites in this study. The red circled sites are from the literatures (NIRS, 2010; Zheng and Yamada, 2005; Zheng and Yamada, 2006b; Zheng and Yamada, 2006c). The surface sediment Pu concentration range was between 0.01–5.81 $mBq\,g^{-1}$.

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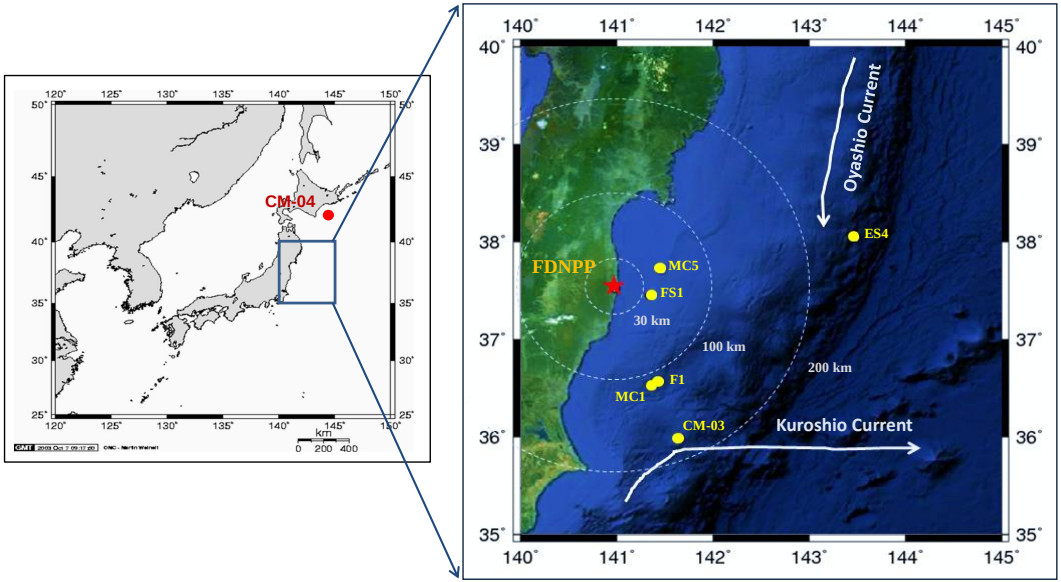

Fig. 2. Map showing the locations of the sediment sampling sites in the western North Pacific and the Kuroshio and Oyashio Currents. Stations CM-04 and CM-03 are from Zheng and Yamada (2006b).

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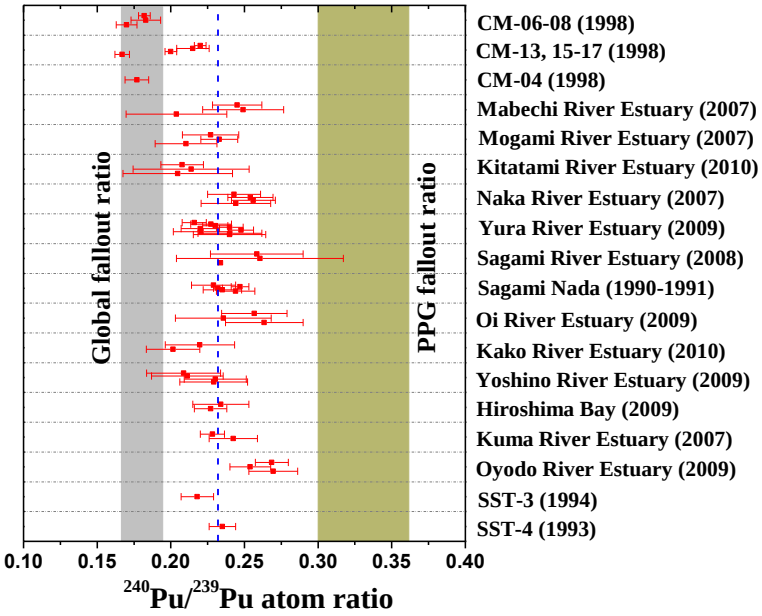

Fig. 3. The $^{240}Pu/^{239}Pu$ atom ratio distribution in Japanese near-coast surface sediments. The years shown in the brackets are the sampling dates. The areas shaded in grey and light green represent the $^{240}Pu/^{239}Pu$ atom ratio ranges of globall fallout (0.180 ± 0.014) and PPG close-in fallout (0.30–0.36), respectively. The atom ratio ranged from 0.170–0.270. For the eastern esturies facing the North Pacific, the average $^{240}Pu/^{239}Pu$ atom ratio was 0.231 ± 0.025 ($n = 36$). The blue dotted line is the mean atom ratio value (0.231). Data for CM-06-08, CM-13, 15-17, Mabechi River Estuary, Mogami River Estuary, Yura River Estuary, Kuma River Estuary, SST-3 and SST-4 are cited from the literatures (NIRS, 2010; Zheng and Yamada, 2005; Zheng and Yamada, 2006b; Zheng and Yamada, 2006c).

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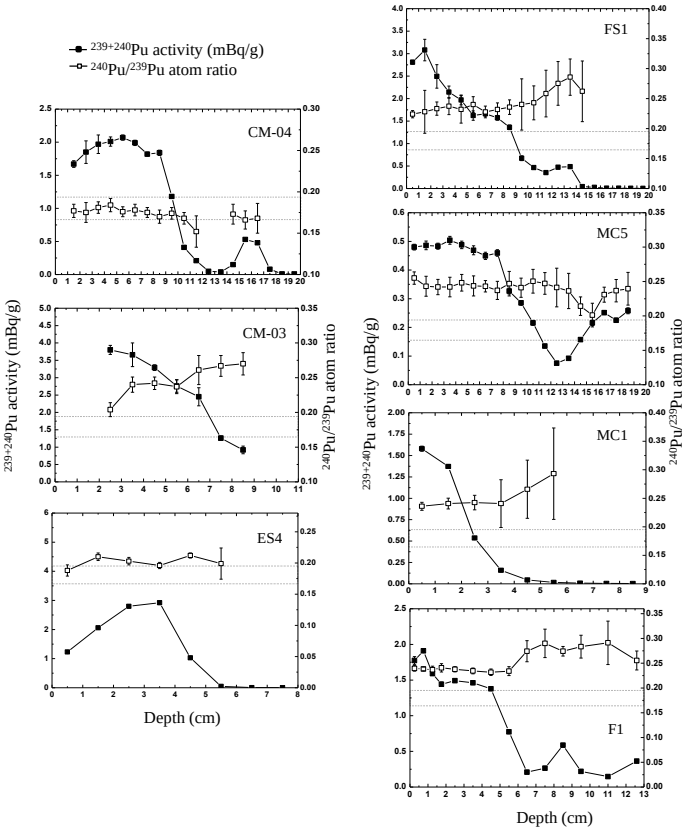

Fig. 4. Vertical profiles of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in the sediment cores. The dashed lines indicate the ^{240}Pu/^{239}Pu atom ratio range of global fallout (0.180 ± 0.014). CM-03 and CM-04 are from Zheng and Yamada (2006b).

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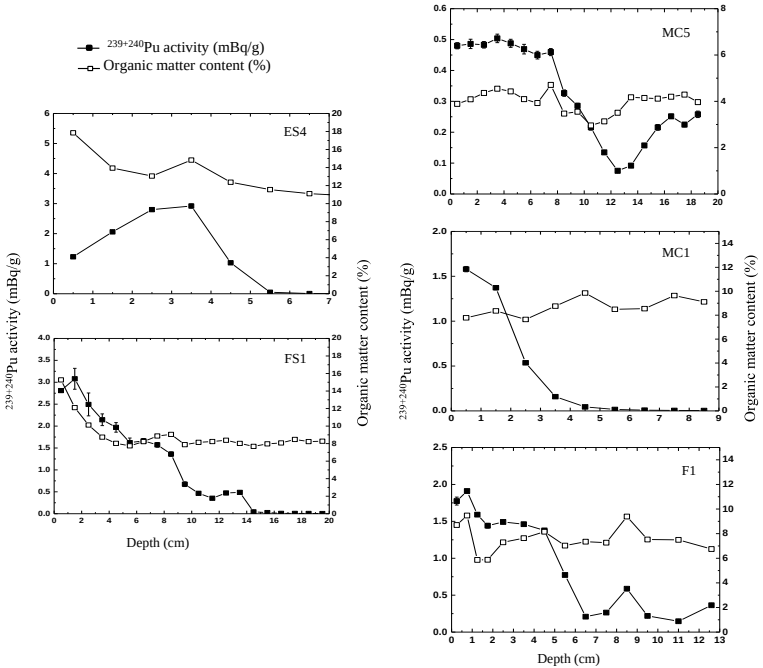

Fig. 5. The vertical profiles of $^{239+240}$Pu activity and organic matter content in the sediment core samples.

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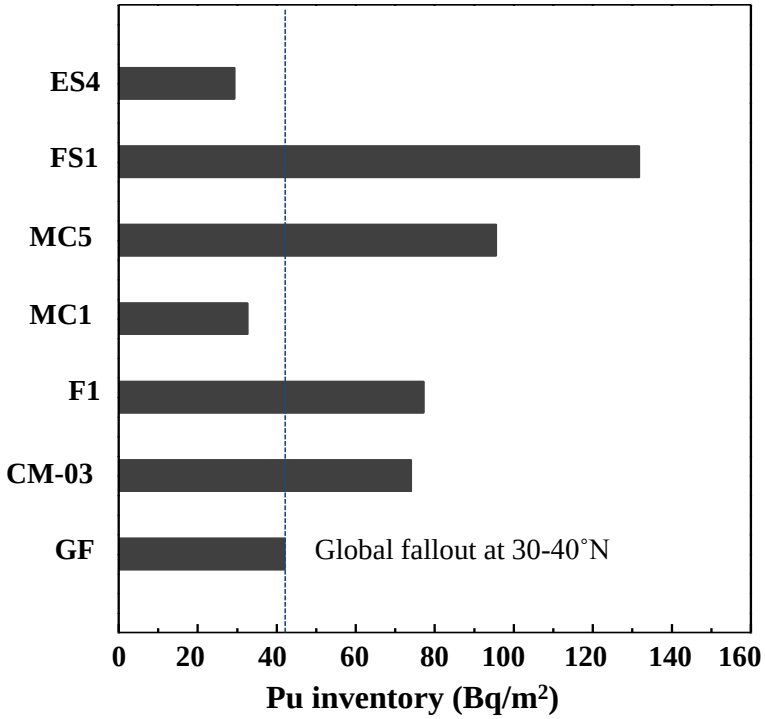

Fig. 6. Pu inventories in the sediment cores. The Pu inventory of CM-03 (0–10 cm) was cited from Zheng and Yamada (2006b). The global fallout Pu inventory at 30–40° N was from UNSCEAR (1993).

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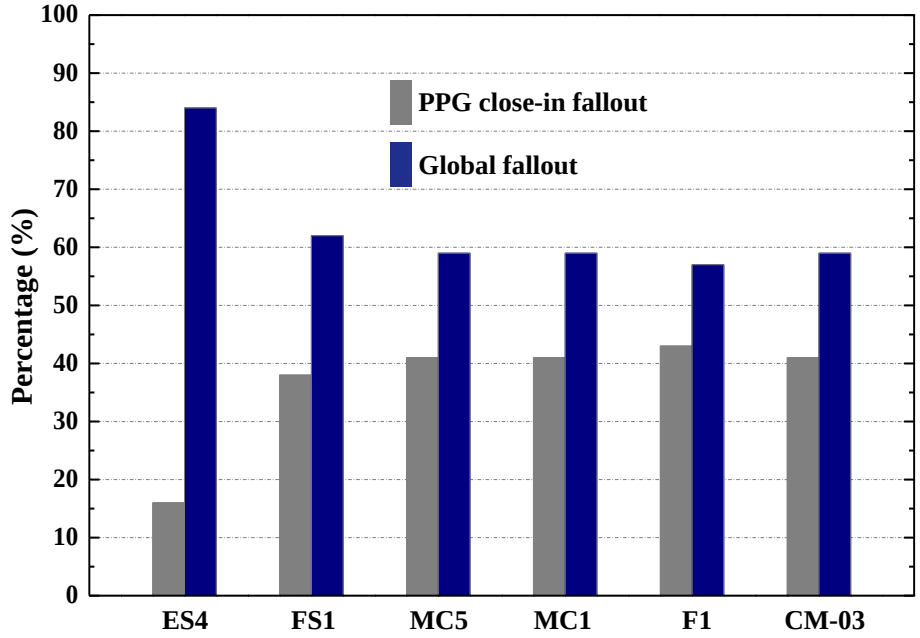

Fig. 7. The percentage of Pu that originated from global fallout and PPG close-in fallout in the sediment cores. The data for CM-03 were calculated from Zheng and Yamdada (2006b).

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