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Temporal distribution of plutonium isotopes in marine sediments off Fukushima after the Fukushima Dai-ichi Nuclear Power Plant accident

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Abstract The characterization of Pu isotopes in a sediment core collected in the western North Pacific off Fukushima in January 2013 was determined. We compared the results with that of the marine sediments collected from July 2011 to July 2012. Both the $^{239+240}$Pu activities and the ^{240}Pu/^{239}Pu atom ratios showed no variation in the marine sediments in the last 2 years since the FDNPP accident. Pu contamination in the marine sediments in the investigated areas still originated from global fallout and the Pacific Proving Ground close-in fallout.

Keywords FDNPP accident · Plutonium isotopes · Marine sediments · SF-ICP-MS

Introduction

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident caused the release of large amounts of radionuclides into the atmosphere as well as into the sea. The radionuclide contamination in the marine environment due to the FDNPP accident is of great public and scientific concern. Investigating the immediate and continuous records of radionuclides in the marine environment off Fukushima after the FDNPP accident is important for source term estimation and understanding the migration behaviors of the radionuclides. The spatial and temporal distribution of radiocesium (^{134}Cs and ^{137}Cs) in the western North Pacific off the FDNPP site after the nuclear accident have been studied intensively [1–3]. However, information about the spatial and temporal distribution of Pu isotopes in the marine environment after the nuclear accident is limited.

The atmospheric released Pu isotopes from the FDNPP accident, characterized by a high ^{240}Pu/^{239}Pu atom ratio of 0.303–0.330, have been detected in litter and soil samples collected around the FDNPP site and the total released amount of $^{239+240}$Pu was estimated to be 1.0–2.4 $\times$ 10^9 Bq [4, 5]. For the marine environment, in the previous works, we investigated the distribution of Pu isotopes in several sediment core samples in the western North Pacific 30 km off the FDNPP site collected from July 2011 to July 2012 and found no detectable Pu contamination from the nuclear accident in the investigated areas [6, 7]. The Pu contamination in the marine sediments 30 km off Fukushima originated from the global fallout and the Pacific Proving Ground (PPG) close-in fallout. It has been modeled that if the discharge of Pu occurred from the FDNPP, it would remain in the near coastal areas [8]. However, to date, no information about the Pu isotopes in the discharged radioactive liquids from the FDNPP accident and the near costal marine environment (within 30 km) is available, thus Pu contamination in the marine environment off Fukushima needs long-term and continuous investigation.

In this work, we determined vertical distribution of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in a sediment core collected off Fukushima in January 2013 to study the temporal distribution of Pu isotopes in the marine

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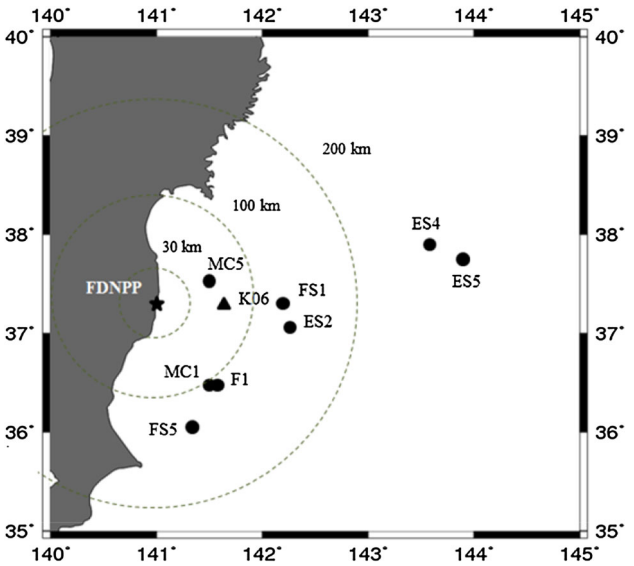

Fig. 1 Sampling stations of the sediments collected in the western North Pacific off the FDNPP site after the nuclear accident. Stations of ES4, ES5, MC5, FS1, ES2, FS5, MC1 and F1 are redrawn from [6, 7]

sediments. Possible sources of Pu isotopes in the marine sediments off Fukushima will be examined and discussed.

Experimental

A sediment core sample (core length 10 cm) was collected by a multiple corer at K06 station (37°20.00′N, 141°40.10′E, water depth 300 m) in the western North Pacific about 100 km off the FDNPP site during the cruise of KT 13-1 in January 2013. The sampling location of K06 together with the locations of sediment core samples collected from July 2011 to July 2012 was shown in Fig. 1.

For the analysis of Pu isotopes, the dried and weighed sediment sample (about 1.5 g) was digested with 20 mL conc. HNO_3 in a tightened Teflon vessel after spiked with 1 pg ^{242}Pu as yield monitor. Then a two-stage anion-exchange chromatographic method using AG 1 × 8 and AG MP-1 M resins was employed for the separation and purification of Pu. This method is described in detail in another work [9]. The chemical recovery of Pu was about 64 % by using this method. Pu isotopes were measured by a double-focusing SF-ICP-MS (Element 2, Thermo Finnigan, Germany).

Results and discussion

Vertical distribution of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in K06

The analytical results of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in the sediment core of K06 were presented in Table 1. The vertical distribution of Pu isotopes at K06 station together with that at MC5 station was shown in Fig. 2. The $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios of K06 ranged from 0.23 to 1.41 mBq g^{-1} and from 0.233 to 0.253, respectively.

Table 1 Results of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in the sediment core at K06 station

Sample depth (cm)	$^{239+240}Pu$ activity (mBq g^{-1})	$^{240}Pu/^{239}Pu$ atom ratio
0–1	0.59 ± 0.02	0.244 ± 0.007
1–2	0.64 ± 0.03	0.247 ± 0.013
2–3	0.61 ± 0.02	0.241 ± 0.015
3–4	0.45 ± 0.01	0.244 ± 0.007
4–5	1.41 ± 0.03	0.239 ± 0.009
5–6	1.09 ± 0.03	0.237 ± 0.008
6–7	0.54 ± 0.02	0.238 ± 0.014
7–8	0.44 ± 0.02	0.233 ± 0.012
8–9	0.35 ± 0.02	0.248 ± 0.018
9–10	0.23 ± 0.01	0.253 ± 0.012

The $^{239+240}Pu$ activity peaked at the depth of 4–5 cm. The subsurface maximum $^{239+240}Pu$ activities have also been reported in other sediment cores in the western North Pacific and its marginal seas [10, 11]. Normally, anthropogenic radionuclides in the sediments of costal and slope areas with a high sedimentation rate showed a subsurface maximum peak that corresponding to 1963 [12]. Kallel et al. [13] reported a sedimentation rate of 0.208 cm $year^{-1}$ in the coastal area of northeastern Japan (41°44′N, 142°33′E). Taking this sedimentation rate into account, the $^{239+240}Pu$ activity maximum at K06 station could be under the 10 cm sediment layer, deeper than that we observed at K06 station. However, sedimentation mixing processes controlled by bioturbation and/or physical factors also affected the vertical distribution of Pu isotopes in marine sediment cores. For example, at MC5 station (shown in Fig. 1), which is close to K06, we observed nearly uniform distribution of $^{239+240}Pu$ activities from the surface to a depth of 8 cm [7]. Thus the cause of the subsurface $^{239+240}Pu$ activity maximum at K06 station needs further investigation. The inventory of $^{239+240}Pu$ at K06 station is 68.9 Bq m^{-2} with a depth of 10 cm.

Unlike the distribution of $^{239+240}Pu$ activities, the $^{240}Pu/^{239}Pu$ atom ratios in the sediment core stayed almost constant with an inventory-weighed value of 0.242. This value is significantly higher than the global fallout value (0.180), clearly indicating the existence of additional sources of Pu. However, this high $^{240}Pu/^{239}Pu$ atom ratio could not be simply attributed to the influence of the FDNPP accident. The PPG-derived Pu, which is characterized by high $^{240}Pu/^{239}Pu$ atom ratios (>0.30), widely existed in the western North Pacific before the FDNPP

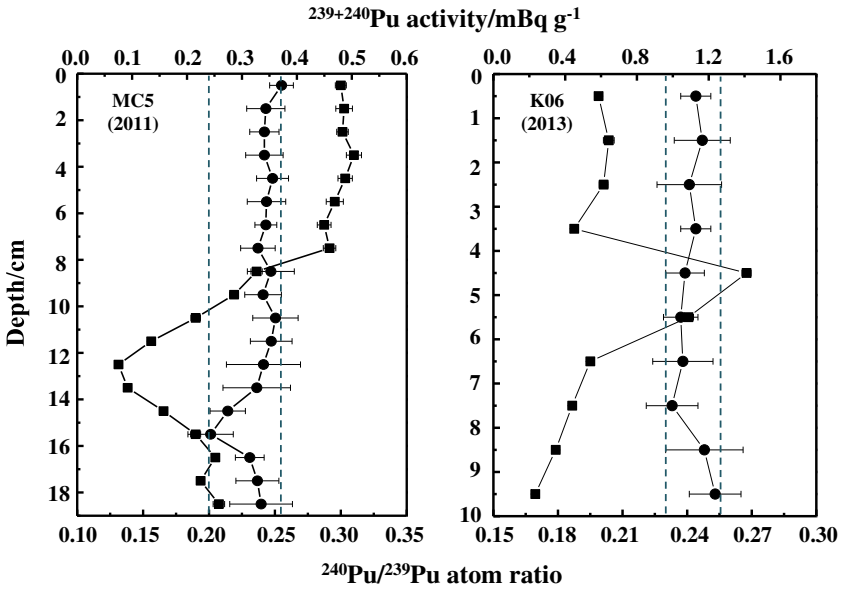

Fig. 2 Vertical distribution of $^{239+240}$Pu activities (*squares*) and ^{240}Pu/^{239}Pu atom ratios (*circles*) in the sediment cores collected at MC5 and K06 stations. The *dashed lines* represent the ^{240}Pu/^{239}Pu atom ratio ranges. Data for MC5 are cited from [7]

Table 2 Pu distribution in the sediments collected in the western North Pacific off the FDNPP site after the nuclear accident from July 2011 to January 2013

Sample	Sampling time	Location	Water depth (m)	Surface $^{239+240}$Pu activity (mBq g^{-1})[a]	Vertical ^{240}Pu/^{239}Pu atom ratio range	$^{239+240}$Pu inventory (Bq m^{-2})	Ref.
MC1	18 July 2011	36°28.97′N, 141°29.93′E	1,327	1.58 ± 0.03	0.236–0.239	32.6 (0–9 cm)	[6, 7]
MC5	19 July 2011	37°35.01′N, 141°30.95′E	141	0.48 ± 0.01	0.201–0.255	95.5 (0–19 cm)	[6, 7]
ES4	18 July 2011	37°51.96′N, 143°34.52′E	5,253	1.23 ± 0.03	0.188–0.212	29.3 (0–8 cm)	[6, 7]
ES5	18 July 2011	37°47.69′N, 143°51.93′E	7,047	1.08 ± 0.03	0.189 ± 0.013[b]	–	[6]
FS1	2 August 2011	37°19.97′N, 142°10.05′E	994	2.81 ± 0.04	0.224–0.286	131.7 (0–20 cm)	[6, 7]
ES2	2 August 2011	37°3.98′N, 142°15.02′E	2,138	3.09 ± 0.04	0.216–0.247	141.4 (0–11 cm)	[7,9]
FS5	3 August 2011	36°00.00′N, 141°20.14′E	1,198	3.53 ± 0.10	0.230 ± 0.015[b]	–	[6]
F1	7 July 2012	36°29.09′N, 141°30.01′E	1,322	1.77 ± 0.05	0.232–0.291	77.2 (0–13.2 cm)	[7]
K06	13 January 2013	37°20.00′N, 141°40.10′E	300	0.59 ± 0.02	0.233–0.253	68.9 (0–10 cm)	This study

[a] Surface sediment 0–1 cm

[b] Only surface sediments were determined

accident [10, 14]. It has been summarized that the ^{240}Pu/^{239}Pu atom ratios in the upper layer (<5 cm) sediments in the western North Pacific before the FDNPP accident could be as high as 0.28 [7]. The ^{240}Pu/^{239}Pu atom ratios of K06 were typically in the background data range. The uniform distribution of ^{240}Pu/^{239}Pu atom ratios from the surface to the depth of 10 cm in the sediment core at K06 station indicated the sufficient mixing of the global fallout Pu and the PPG close-in fallout Pu.

Temporal distribution of Pu isotopes in marine sediments after the FDNPP accident

The information about the Pu characterization in all the sediments was summarized in Table 2. The surface $^{239+240}$Pu activity of K06 was significantly lower than that of all the other stations except MC5. MC5 and K06 are the two closest stations to the FDNPP site and the $^{239+240}$Pu activities in the surface sediments of these two stations were similar (0.48 ± 0.01 mBq g^{-1} for MC5 and 0.59 ± 0.02 mBq g^{-1} for K06). The $^{239+240}$Pu inventory at K06 station is slightly lower than that of MC5. As the sampling time of K06 is one and a half year later than that of MC5, these results suggested that there has been no variation for Pu distribution in the marine sediments in that region during the last 2 years after the FDNPP accident.

For the sediment cores collected from July 2011 to July 2012, the ^{240}Pu/^{239}Pu atom ratios ranged from 0.188 to 0.291. The ^{240}Pu/^{239}Pu atom ratios in the deeper layers (>6 cm) of FS1 and F1 could be higher than 0.27 due to the

deposition of PPG close-in fallout Pu at the early stage of the explosion of the nuclear tests [7]. The $^{240}Pu/^{239}Pu$ atom ratios in the sediment core at K06 station were typically in the range of other stations. These results suggested that the sources of Pu contamination in the marine sediments 30 km off the FDNPP site remained constant during the last 2 years after the FDNPP accident and the Pu isotopes originated from global fallout and the PPG close-in fallout.

Resolving the sources of Pu contamination

By using a simple two end-member mixing model, we estimated the contributions of the two sources (global fallout and PPG close-in fallout) for the Pu distribution in the marine sediments. Presuming the $^{240}Pu/^{239}Pu$ atom ratio for global fallout is 0.18 and the $^{240}Pu/^{239}Pu$ atom ratio for PPG close-in fallout is 0.36, the inventory-weighted percentage of the PPG close-in fallout Pu in the sediments at K06 station was calculated to be 42 %. This result was consistent with that (38–43 %) of the other four stations (MC1, MC5, FS1, F1), but higher than that (16 %) of the ES4 station [7]. In the marginal seas of the western North Pacific, such as the Sagami Bay, the South China Sea and the Hiroshima Bay, researchers also observed about 40 % contribution of the PPG close-in fallout Pu to the Pu contamination in the marine sediments before the FDNPP accident [10, 15, 16].

Conclusions

In this paper, we determined the vertical distribution of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in a sediment core collected 30 km off the FDNPP site in January 2013 to investigate the temporal distribution of Pu isotopes in marine sediments after the FDNPP accident. Both the $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios showed no variation in the marine sediments for the last 2 years after the FDNPP accident. Global fallout and the PPG close-in fallout are the two sources for the Pu contamination in the marine sediments.

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