Investigating plutonium contamination in marine sediments off Fukushima coast following the Fukushima Dai-ichi Nuclear Power Plant accident

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident has caused large amounts of anthropogenic radionuclides to be released into the atmosphere as well as directly discharged into the sea. To obtain the vertical distribution of Pu isotopes in marine sediments and to better assess the possible contamination from the FDNPP accident in the marine environment, activities of $^{239+240}$Pu and ^{241}Pu, as well as the atom ratios of ^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu, were investigated in a sediment core collected from the western North Pacific in July 2011. The observed vertical profile of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios showed no extra injection of Pu from the accident, indicating no immediate Pu contamination from the FDNPP accident in the marine sediments in the region investigated.

Key Words: *FDNPP, plutonium isotopes, marine sediments, SF-ICP-MS*

1. Introduction

On March 11, 2011, the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) sustained severe damage due to a magnitude 9.0 earthquake in the northwest Pacific about 130 km off the northeast coast of Japan and the ensuing gigantic tsunami. As a result, the cooling system of the nuclear reactors failed, leading to hydrogen explosions inside the reactors. In addition to atmospheric releases caused by the hydrogen explosions and venting activities, highly contaminated water with large amounts of radionuclides, due to desperate attempts to prevent reactor cores meltdowns by injecting water into the reactors, was directly discharged or leaked into the North Pacific Ocean[1)].

Intensive studies on the released high volatility fission products in 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. However, studies focusing on the possible released actinides, especially Pu isotopes, are limited. Pu isotopes are characterized by high chemical toxicity and radiotoxicity, and they have attracted increasing scientific and public concern[2)]. Thus, it is essential to investigate the impact of the FDNPP accident on the distribution of Pu isotopes in the environment. Abnormal atom or activity ratios of ^{240}Pu/^{239}Pu, ^{241}Pu/$^{239+240}$Pu and ^{238}Pu/$^{239+240}$Pu were reported in the surface soils in the 20–30 km zones around the FDNPP, providing evidence of the atmospheric release of Pu[3),4)]. For the marine environment, in a previous work[5)], we investigated the distribution of Pu isotopes in surface marine sediments collected several months after the accident, from the Pacific Ocean, 30 km off the FDNPP, and we observed no significant variation in $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio compared with

those previously reported in marine sediments in the western North Pacific and its marginal seas. Meanwhile, Pu concentration in seawaters in the Pacific, 50 km off the FDNPP, showed no extra components from the accident[6]. However, the possible long-term Pu contamination in the marine environment remains unknown as no information on the Pu isotopes in the released radioactive liquid and in the FDNPP near the coastal (within 30 km) marine environment is currently available. The radiocaesium (^{137}Cs and ^{134}Cs) originating from the accident was detected in the suspended 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[7]. In addition, Pu isotopes in the western North Pacific can be transported by oceanic currents, e.g., the North Equatorial Current and the Kuroshio Current, flowed by advection and mixing[8]. Thus the possible oceanic Pu contamination in terms of inventory and range from the FDNPP accident needs an on-going investigation before a reliable conclusion can be reached.

In this work, we investigated the vertical distribution of Pu activities and Pu atom ratios (^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu) in one sediment core collected after the accident that occurred northeast of Fukushima to obtain information on Pu inventory in the sediment column, the temporal variation of Pu activities, and isotopic composition.

2. Materials and Methods

A sediment core sample was collected by a multiple corer at ES4 station (37° 53' N; 143° 35'E) in the Pacific about 200 km off the FDNPP during the cruise of KH 11-07 in July 2011. The water depth was 5400 m. The location of the sampling site is shown in Fig. 1.

Sediment samples were digested with concentrated HNO_3 after they were spiked with ^{242}Pu (ca. 1 pg). A two-stage anion-exchange chromatographic method was employed for the separation and purification of Pu[9]. Pu isotopes were measured by a double-focusing sector field ICP-MS (Element 2, Thermo Finnigan, Bremen, Germany). The operation conditions and measurement parameters for this analytical method have been described previously[10].

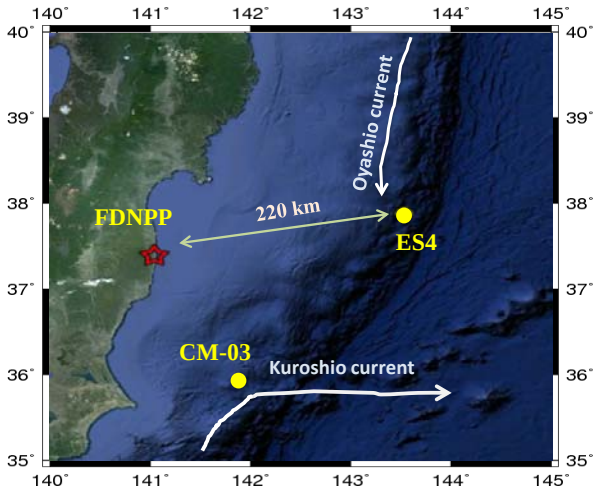

Fig. 1. Map showing the sampling station and the Kuroshio and Oyashio Currents. Station CM-03 is redrawn from ref. 11.

3. Results and Discussion

(1) Vertical distribution of Pu activities

The analytical results for Pu isotopes are presented in Table 1 and Fig. 2. The $^{239+240}$Pu activities ranged from 0.01 to 2.92 mBq/g, while the ^{241}Pu activity was only detectable with a value of 7.42 mBq/g in the sediment at a depth of 3–4 cm where $^{239+240}$Pu activity peaked. The surface sediment Pu concentration was previously compared with the baseline data of the adjacent open oceans of the Pacific Ocean; it showed no significant increase[5]. The highest $^{239+240}$Pu activity (2.92 mBq/g) in ES4 sediment core was lower than that (3.80 mBq/g) at CM-03, which is located ca. 200 km southeast of the FDNPP with known Pu vertical distribution information before the accident[11].

Moon et al. (2003)[12] reported relatively low sedimentation rates (0.12–0.35 cm/ky) in the Northwest Pacific Basin after they investigated the ^{14}C and ^{232}Th$_{ex}$ distribution 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 in that region. However, the biological activity in the sea floor in the western North Pacific is high, evidencing a high bioturbation rate in the surficial marine sediment[13]. The particle mixing coefficient ranges from 0.02 to 1.00 cm^2/y in the western North Pacific[12]. If we assume that 48 years 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 a calculation from the radiochemical model of the sedimentary process described by Guinasso and Shink (1975)[14], which is consistent with the result we observed at

Table 1. Results of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in the sediment core at ES4 station.

Depth (cm)	Dry bulk density (g/cm^3)	$^{239+240}$Pu activity (mBq/g) dry weight	^{240}Pu/^{239}Pu atom ratio
0-1[a]	0.32	1.23±0.03	0.188±0.009
1-2	0.27	2.06±0.04	0.210±0.006
2-3	0.30	2.80±0.05	0.203±0.006
3-4	0.25	2.92±0.06	0.196±0.005
4-5	0.38	1.03±0.04	0.212±0.004
5-6	0.33	0.05±0.01	0.198±0.025
6-7	0.48	0.01±0.00	ND

[a] Data for 0-1 cm are cited from ref. 5; ND: not detected.

ES4. Similarly, Harada and Shibamoto (2002)[13] observed that the penetration depth of excess ^{210}Pb was 7.2cm in the same sea area of ES4.

The plutonium inventory at ES4 was calculated to be 29 Bq/m^2, which was slightly lower than the total cumulative global fallout value (42 Bq/m^2)[15]. In the adjacent sea area of ES4, Moon et al. observed nearly identical results (28.6 Bq/m^2) in sediment cores collected in 1997[12], suggesting no significant amount of Pu injection into the sediment in that region in the past 15 years.

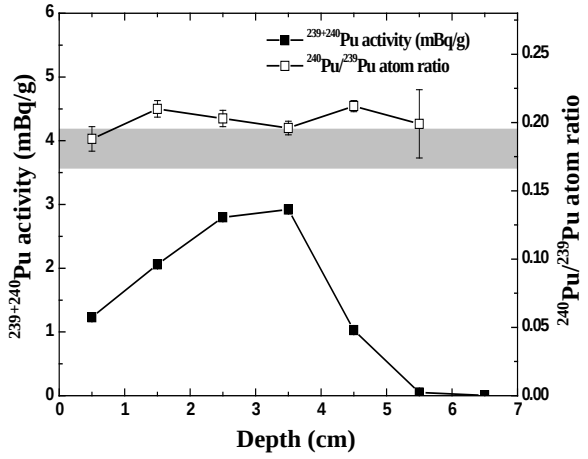

Fig. 2. Vertical distribution of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio at ES4. The area shaded in grey represents global fallout atom ratio ranges (0.180 ± 0.014, 30–70° N)[16].

(2) Pu atom ratios and possible sources of Pu

Pu atom ratios (^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu) have been demonstrated to be good indicators for Pu source identification, as higher ratios can be expected with the higher neutron fluxes associated with an increase in the yield of a nuclear detonation[17]. Atom ratios of ^{240}Pu/^{239}Pu (0.303–0.330) and ^{241}Pu/^{239}Pu (0.103–0.135), which were higher than global fallout values, were found from the analysis of Pu isotopes in the surface soil and litter samples in the 20–30 km zones around the FDNPP[3]. For the western North Pacific, Pu in the sediments could be attributed to the global fallout and the Pacific Proving Ground (PPG) close-in fallout before the FDNPP accident. It was summarized in our previous work that the ^{240}Pu/^{239}Pu atom ratios in the western North Pacific and its marginal seas before the FDNPP accident ranged from 0.150 to 0.281[5].

The ^{240}Pu/^{239}Pu atom ratios in the ES4 sediment core ranged from 0.188 to 0.212, with the inventory-weighted mean of 0.202. The observed results were typically in the background value range, indicating the mixing of global fallout Pu and PPG close-in fallout Pu. The ES4 station is located in the pathway of the southward flowing Oyashio Current, which carries water-bearing global fallout Pu isotopes. Traces of the PPG derived Pu could be from direct fallout in that region when the nuclear weapon tests were conducted. For the ^{241}Pu/^{239}Pu atom ratio (for all the values discussed here, decay corrections have been made to 1 January 2000), at the depth of 3–4 cm, a value of 0.0050 was observed, which is higher than the global fallout value (0.0019)[16]. However, this result was similar to the result (0.0061) we observed in the sediment at ES2[5], and is nearly consistent with the results (0.0053-0.0057) for the sediment near the Bikini Atoll reported by Lee et al.[8]; therefore, this is further evidence of the PPG source contribution to that region. Thus, it could be concluded that the Pu isotopes at the ES4 station originated mainly from the global fallout and the trace of the PPG close-in fallout, and that there was no extra Pu injection from the FDNPP in that region 4 months after the accident. Presuming that the ^{240}Pu/^{239}Pu atom ratio for global fallout is 0.18[16], and that the representative ^{240}Pu/^{239}Pu atom ratio for the PPG close-in fallout is 0.36[18], the PPG source contribution is calculated to be 16% by using a two-end member–mixing model.

4. Conclusions

The vertical distribution of Pu isotopes in the sediment at the ES4 station in the western North Pacific that was collected in July 2011 was investigated. The inventory of $^{239+240}$Pu activity and the inventory-weighted ^{240}Pu/^{239}Pu atom ratio were 29 Bq/m^2 and 0.202, respectively, showing no extra injection of Pu isotopes from the FDNPP accident. The global fallout and the PPG close-in fallout were the two sources of Pu contamination in that region, and the bioturbation effect was shown to be the key factor that controlled the Pu vertical profile. Although Pu originating from the accident was not observed in the ES4 sediment samples, it should be noted that the possible long-term Pu contamination from the FDNPP accident in the marine environment needs further investigation before a comprehensive assessment can be achieved.

Acknowledgments

This work was supported by MEXT (24110004), and partly supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan.

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