Release of plutonium isotopes from the Fukushima Daiichi Nuclear Power Plant accident

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The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident has caused serious contamination in the environment. In this work, we summarize and analyze published studies related to the release of Pu from the FDNPP accident using environmental sample analysis and ORIGEN model simulation. Our analysis focuses on several aspects: first, the investigation of the distribution of Pu isotopes derived from the FDNPP accident in the environment; second, the determination of Pu isotopic composition of the FDNPP-derived Pu in the environment; third, the identification of sources of Pu release in the FDNPP-damaged reactors or spent fuel pools; and finally, the estimation of the amount of Pu isotopes released from the FDNPP.

Key Words : *Plutonium, Sources, Isotopic composition, FDNPP*

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident has caused serious contamination in the environment from the atmospheric fallout and the direct discharges of highly radioactive liquid wastes.[1)-3)] In addition to the massive release of highly volatile fission products, such as ^{127m}Te, ^{132}Te, ^{131}I, ^{133}Xe, ^{134}Cs, ^{136}Cs, and ^{137}Cs, the possible release of non-volatile Pu isotopes attracted considerable public attention following the FDNPP accident. It is known that Pu isotopes present a large risk for internal radiation exposure via ingestions of contaminated agricultural crops, particularly for ^{241}Pu (a beta-emitter, $T_{1/2}$ = 14.4 years). As it decays, the ingrowth of ^{241}Am (an alpha and gamma-emitter, $T_{1/2}$ = 432.7 years) will present an additional radiation risk.

Plutonium is generally produced in reactor fuel as a mixture of isotopes. The predominant isotope, ^{239}Pu, is produced by neutron capture by ^{238}U. If a fuel element containing ^{239}Pu is left in a reactor for any length of time, further neutron capture can occur, yielding isotopes with higher mass such as ^{240}Pu, ^{241}Pu, and ^{242}Pu. In addition, small quantities of two other isotopes, ^{236}Pu and ^{238}Pu, are produced during the irradiation.[4)] To date, information on the release of Pu isotopes from the FDNPP accident to the environment is very limited.

Several studies on the determination of Pu isotopes in environmental samples, such as soil, litter, sediment, seawater, and aerosols, confirmed the release of Pu isotopes from the FDNPP accident.[5)-10)] Recently, core inventory calculation results of Pu isotopes within the FDNPP reactors using the ORIGEN model simulation have been reported.[11)-12)] These studies suggest that the damaged reactors were the major contributor of Pu to the environment. However, the release of Pu from the spent fuel pools remains unknown. In this work, we summarize and analyze the published studies related to the release of Pu isotopes from the FDNPP accident. Our analysis will focus on: (1) the distribution of Pu isotopes derived from the FDNPP accident in the environment; (2) Pu isotopic composition for source identification; (3) sources for Pu release in the FDNPP-damaged reactors or spent fuel pools; and (4) the amounts of Pu isotopes released from the FDNPP accident. The paper concludes with recommendations for future research directions.

2. Release of Pu isotopes from the accident

(1) Distribution of Pu isotopes derived from the FDNPP accident in the environment

According to the air dose monitoring data from the MEXT (Ministry of Education, Culture, Sports, Science and Technology) and the atmospheric dispersion simulation by SPEEDI, a high concentration plume of the released radionuclides moved towards the northwest from the power plant during the daytime on March 15, 2011; consequently, a large amount of radionuclides were deposited on the ground by precipitation.[13] Investigation conducted by the MEXT on the distribution of Pu isotopes in surface soils in Fukushima prefecture found no significant increase of $^{239+240}$Pu activity in soil samples, however, ^{238}Pu/$^{239+240}$Pu activity ratios (0.33–2.2) higher than that of the global fallout (0.026) were detected at five sites, indicating possible distribution of Pu related to the FDNPP accident northwest of the FDNPP.[14] Similarly, using the alpha counting technique, Yamamoto et al.[5)-6)] completed a field survey soon after the accident in the heavily contaminated areas outside the 20 km exclusion zone, as well as in Okuma Town adjacent to the plant. The anomaly of the ^{238}Pu/$^{239+240}$Pu activity ratios (0.059–1.324) indicated the existence of trace amounts of Pu isotopes originating from the accident in soils from Iitate Village and Okuma Town. These studies, however, did not provide more information on Pu isotopic composition in the environmental samples. In particular, ^{241}Pu, the principle isotope contributing to the dose due to external exposure from radioactivity deposition after the accident, was not considered in the investigations.

The release of ^{241}Pu from the FDNPP accident was confirmed for the first time by Zheng et al.[7] They analyzed Pu activity ($^{239+240}$Pu and ^{241}Pu) and its isotopic composition (^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu atom ratios) using a sector-field ICP-MS (SF-ICP-MS) in surface soil and litter samples collected in Fukushima prefecture in 20–30 km zones (sampling in April and May 2011) and soil samples collected in Mito City, Kamagaya City, and Chiba City (sampling in April–August 2011). For the samples collected in Fukushima Prefecture, activities of $^{239+240}$Pu ranged from 0.019 to 1.400 mBq/g, within the typical global fallout $^{239+240}$Pu activity range of 0.15 to 4.31 mBq/g observed in Japanese soils before the Fukushima DNPP accident.[15] However, high activities of ^{241}Pu ranging from 4.52 to 34.8 mBq/g were detected in samples of the J-Village surface soil (0–2 cm) and in litter at sites S2 and S3 (Fig. 1).

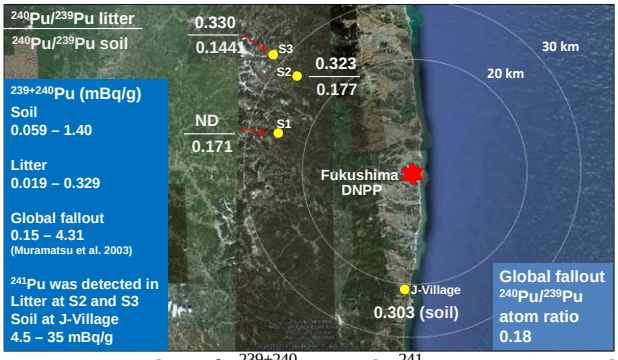

Fig. 1. Results of $^{239+240}$Pu and ^{241}Pu activities and ^{240}Pu/^{239}Pu atom ratios in surface soil and litter samples collected in the Evacuation-Prepared Area (J-Village) and the Deliberate Evacuation Area (S1, Katsurao Village; S2, Namie Town; and S3, Iitate Village).

^{241}Pu was released into the environment during atmospheric nuclear weapons testings in the last century. Because of its short half-life of 14.4 years, the activity of ^{241}Pu in Japanese soils before the FDNPP accident was quite low (ca. 1.2 for ^{241}Pu/$^{239+240}$Pu activity ratio, ^{241}Pu decay corrected to March 15, 2011). Therefore, the finding of high ^{241}Pu activities in these samples suggested an additional Pu input. The ^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu atom ratios found in these samples ranged from 0.303 to 0.330 and from 0.103 to 0.135, respectively. They were significantly higher than those of global fallout (0.180±0.007, 1σ for ^{240}Pu/^{239}Pu atom ratio, and 0.00194±0.00014, 1σ for ^{241}Pu/^{239}Pu atom ratio)[16] and the atmospheric fallout deposition in Japan from 1963 to1979 (0.1922±0.0044, 1σ for ^{240}Pu/^{239}Pu atom ratio, and 0.00287±0.00056, 1σ for ^{241}Pu/^{239}Pu atom ratio),[17] indicating new Pu input from the Fukushima DNPP accident. We noted that in the surface soil (0–1cm) under the litter layer at sites S3 and S2, no ^{241}Pu was detected and ^{240}Pu/^{239}Pu atom ratios were 0.144 and 0.177, respectively, close to the global fallout value of 0.180. This phenomenon indicated that the released Pu, deposited in the litter layer, had not reached the underlying surface soil by May 2011, when the samples were collected. No ^{241}Pu could be detected in surface soils in Mito, Kamagaya and Chiba Cities, located 100–200 km away from the FDNPP; in addition, atom ratios of ^{240}Pu/^{239}Pu in these samples showed typical global fallout value, indicating that the Fukushima-sourced contribution to the total Pu activity in these areas was negligible. However, long-distance transport of Fukushima-sourced Pu was suggested by

Lujaniene et al.[8] They observed an elevated $^{238}Pu/^{239+240}Pu$ activity ratio (1.2) and $^{240}Pu/^{239}Pu$ atom ratio (0.244±0.018) in an integrated air sample collected from 23 March to 15 April, 2011 (n = 30) in Vilnius, Lithuania, and suggested 43%–59% Fukushima-derived $^{239+240}Pu$ to the total $^{239+240}Pu$ activity. Obviously, further study is needed to clarify the possible long-distance transport of Fukushima source Pu, as the possibility of contributions of re-suspended Pu from Siberian nuclear plants and deposited Chernobyl accident-derived Pu could not be excluded.

Information on the distribution of Pu isotopes in the marine environment following the FDNPP accident is quite limited. Zheng et al.[9] reported the first dataset on the distribution of Pu isotopes in surface sediments in the Pacific 30 km off Fukushima after the accident. Activities of $^{239+240}Pu$ and ^{241}Pu and atom ratios of $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ in surface sediments collected between July to August 2011 were analyzed to make a quick assessment of the environmental impact of the possible Pu contamination. The observed $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios are comparable to those previously reported in marine sediments in the western North Pacific and its marginal seas, and in Japanese estuaries before the accident. Pu contamination from the Fukushima DNPP accident was not observed in marine sediments outside the 30 -km zone. This conclusion was further supported by Sakaguchi et al.[10] They analyzed Pu isotopes in seawater collected during the same sampling cruise of sediment samples in July–August 2011. The Pu concentration and isotopic composition ($^{240}Pu/^{239}Pu$ atom ratio of 0.181–0.218) were similar to the baseline data before the accident, indicating no immediate Pu contamination in the investigated seawater. However, the contamination situation within the 30 -km zone around the plant remains unknown. Further studies on the determination of Pu isotopes in seawater and sediments within the 30 -km zone are required to make a more comprehensive assessment on the environmental impact of the Fukushima DNPP accident and to understand the marine environmental behavior of Pu isotopes derived from the Fukushima DNPP accident.

(2) Pu isotopic composition for source identification

Accurate determination of Pu isotopic composition in the environment is important for source identification and long-term tracking of the biogeochemical cycles of the FDNPP-sourced Pu in the environment. It may also provide useful information for the estimation of reactor damage, as high radiation levels make it impossible to directly measure damage to the melted reactor cores.

To obtain accurate isotopic composition for the FDNPP-sourced Pu in the environment, the contribution of global fallout Pu has to be kept as low as possible. Considering the fact that no significant increase in $^{239+240}Pu$ activity in soils was found following the accident, high contribution of global fallout Pu in soils could hamper obtaining accurate FDNPP-derived Pu isotopic composition in the environment.

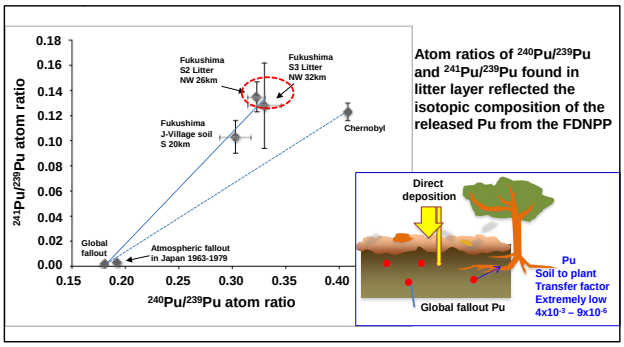

Fig. 2. Isotopic composition of the FDNPP-sourced Pu in forest litter samples in Fukushima Prefecture and a comparison of isotopic composition with those of the Chernobyl accident and the global fallout sources. Redrawn from reference [7].

Zheng et al.[7] attempted to detect the FDNPP Pu signature in litter samples in a forest in Fukushima Prefecture (Fig. 2). It has been well-established that the soil-plant transfer factor of Pu is extremely low (4×10^{-3}–9×10^{-6}),[18] and thus, before the accident, the presence of global fallout Pu in the dead leaves via the soil-plant transfer can be assumed to be negligible. Therefore, we consider that atom ratios of $^{240}Pu/^{239}Pu$ (0.330) and $^{241}Pu/^{239}Pu$ (0.135), and $^{241}Pu/^{239+240}Pu$ activity ratio (108) found in litter samples in the Fukushima Prefecture forest reflected the isotopic composition of Pu isotopes released from the FDNPP accident. As shown in Fig. 2, Pu isotopic composition observed in surface soils of the J-Village was located in the mixing line between global fallout and FDNPP Pu isotopes, indicating 87% contribution of FDNPP Pu and 13% contribution of global fallout to Pu composition.

(3) Sources of Pu release in the FDNPP damaged reactors or spent fuel pools

The three damaged reactor cores contained about 256 metric tons of nuclear fuel. The spent fuel pool in Unit 4 reactor building was the largest one on site, containing

1331 spent nuclear fuel assemblies, which is 1.4 times the amount of fuel loaded within reactors Unit 1 and Unit 3.[12] Therefore, compared to the Chernobyl accident, the FDNPP accident presented a much more complex situation, because both the damaged reactor cores and spent fuel pools may have contributed to the release of Pu isotopes to the environment.

Recently, several groups published model calculation results on the core inventory of Pu isotopes; these studies provided opportunity to identify the sources of Pu release in the accident. Based on the core inventory of Pu isotopes within 1 day after reactor scramming, calculated by Schwantes et al.[12] using an ORIGEN ARP model, Pu isotopic composition of damaged reactor cores can be derived: 0.447, 0.196, 123.7, and 1.92 for $^{240}Pu/^{239}Pu$ atom ratio, $^{241}Pu/^{239}Pu$ atom ratio, $^{241}Pu/^{239+240}Pu$ activity ratio, and $^{238}Pu/^{239+240}Pu$ activity ratio, respectively. A similar ORIGEN ARP module calculation by Kirchner et al.[11] gave lower Pu isotope ratios: 0.395, 0.174, 118.1, and 1.77 for $^{240}Pu/^{239}Pu$ atom ratio, $^{241}Pu/^{239}Pu$ atom ratio, $^{241}Pu/^{239+240}Pu$ activity ratio, and $^{238}Pu/^{239+240}Pu$ activity ratio, respectively. Isotopics group[19] reported $^{240}Pu/^{239}Pu$ atom ratio ranges of 0.30–0.37 for the damaged reactor cores and 0.45–0.60 for the spent fuel pools in the FDNPP. Compared with the Pu isotopic composition observed in litter samples in Fukushima Prefecture,[7] the atom ratio of $^{240}Pu/^{239}Pu$ (0.33) and $^{241}Pu/^{239+240}Pu$ activity ratio (108.7) are in good agreement with the Pu isotopic composition in the damaged reactor cores, indicating that Pu isotopes were released from the damaged reactors, but not from the spent fuel pools in the FDNPP.

(4) The amounts of Pu isotopes released from the FDNPP accident

Unlike the intensive studies on the estimation of total amounts of fission products, such as ^{131}I and ^{137}Cs,[1-4] very few studies have addressed the estimation of total amounts of Pu isotopes released from the FDNPP accident, due partly to the lack of environmental monitoring data.

Zheng et al.[7] made a rough estimation of the atmospheric release of Pu isotopes based on the $^{137}Cs/^{239+240}Pu$ activity ratio observed in forest litter samples in the 20–30 km zones relative to the total amount of ^{137}Cs released estimated by METI[20] and Stohl et al.,[2] with an assumption that ^{137}Cs and Pu siotopes followed the same deposition mechanism, and no significant variation of the $^{137}Cs/^{239+240}Pu$ activity ratio during the release and deposition. As shown in Table 1, the total amounts of released $^{239+240}Pu$ and ^{241}Pu were 1.0×10^{9} – 2.4×10^{9} Bq, and 1.1×10^{11} – 2.6×10^{11} Bq, respectively. These values are very close to those estimated by METI,[20] and about 4 orders of magnitude lower than those of the Chernobyl accident.[21-23]

Table 1. Estimation the amounts of Pu isotopes released from the FDNPP accident and a comparison with Chernobyl.

		Fukushima DNPP	
	Chernobyl	METI calculated	Estimation of Zheng et al.[7]
Amount of released (Bq)			
Pu-239+240	8.7×10^{13}	6.4×10^{9}	1.0×10^{9} – 2.4×10^{9}
Pu-241	7.2×10^{15}	1.2×10^{12}	1.1×10^{11}– 2.6×10^{11}
Pu inventories at reactors (Bq) at the time of accident initiation			
Pu-239+240	2.4×10^{15}	---	8.3×10^{15}
Pu-241	1.9×10^{17}	---	7.0×10^{17}
Percentage of core inventory released (%)			
Pu-239+240	3.5	---	1.2×10^{-5} – 2.9×10^{-5}
Pu-241	3.5	---	1.6×10^{-5} – 3.7×10^{-5}

It was found that although the inventories of Pu isotopes in the reactors in the Fukushima DNPP were ca. 3.5 times higher than those in the Chernobyl Unit 4 reactor,[24] the percentages of core inventory released for both $^{239+240}Pu$ ($1–3\times10^{-5}$ %) and ^{241}Pu ($2–4\times10^{-5}$ %) were about 5 orders of magnitude lower than those of the Chernobyl accident. This estimation is in a good agreement with that of Grambow and Poinssot.[25] However, Schwantes et al.[12] reported an estimation that roughly 2×10^{-3} % of the total plutonium inventory from Units 1 and 3 was released into the environment, which is two orders of magnitude higher than those of Zheng et al.[7] and Grambow and Poinssot.[25] More studies are required to improve the accuracy of estimation of the release of Pu isotopes.

3. Future research directions

Regarding the release of Pu isotopes into the environment from the FDNPP accident, we consider that the following aspects should be addressed in future studies:

(1) Possible Pu contamination in the marine environment due to the direct discharge of highly radioactive liquid waste.

(2) Improvement of the estimation accuracy of the total amounts of Pu isotopes released, and elucidation of the release mechanism.

(3) Because of the high activity ratio of $^{241}Pu/^{239+240}Pu$, the ingrowth of ^{241}Am should be considered in future studies.

(4) Development of highly sensitive Pu particle analytical

techniques to verify possible long-distance transport.

(5) Studies on biogeochemical cycle of Pu isotopes to understand the environmental behavior and fate of the Fukushima-sourced Pu in the environment.

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