Environmental Pollution 163 (2012) 243–247

Contents lists available at SciVerse ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Deposition of fission and activation products after the Fukushima Dai-ichi nuclear power plant accident

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ARTICLE INFO

Article history:
Received 22 August 2011
Received in revised form
24 December 2011
Accepted 1 January 2012

Keywords:
Fukushima Dai-ichi nuclear power plant accident
Fission product
Activation product
^{239}Np

ABSTRACT

The Great Eastern Japan Earthquake on March 11, 2011, damaged reactor cooling systems at Fukushima Dai-ichi nuclear power plant. The subsequent venting operation and hydrogen explosion resulted in a large radioactive nuclide emission from reactor containers into the environment. Here, we collected environmental samples such as soil, plant species, and water on April 10, 2011, in front of the power plant main gate as well as 35 km away in Iitate village, and observed gamma-rays with a Ge(Li) semiconductor detector. We observed activation products (^{239}Np and ^{59}Fe) and fission products (^{131}I, ^{134}Cs (^{133}Cs), ^{137}Cs, ^{110m}Ag (^{109}Ag), ^{132}Te, ^{132}I, ^{140}Ba, ^{140}La, ^{91}Sr, ^{91}Y, ^{95}Zr, and ^{95}Nb). ^{239}Np is the parent nuclide of ^{239}Pu; ^{59}Fe are presumably activation products of ^{58}Fe obtained by corrosion of cooling pipes. The results show that these activation and fission products, diffused within a month of the accident.

1. Introduction

The Great East Japan Earthquake and the subsequent Tsunami on March 11, 2011, caused the cooling systems of the Fukushima Dai-ichi nuclear power plant to break down. The cooling of some of the nuclear reactors thus became impossible, and the pressure in the nuclear reactor container rose because of hydrogen buildup. Despite the venting operation performed for reactor No. 1 on March 12, the building was damaged by a hydrogen explosion. Immediately thereafter, ^{137}Cs was detected from dust collected within the premises of the power plant. Although a venting operation was performed for reactor No. 3 on March 13, a large-scale hydrogen explosion occurred the following day. On the other hand, dry venting was performed for reactor No. 2 without a hydrogen explosion, and a hydrogen explosion occurred in reactor No. 4 on March 15. For these reasons, the nuclear-reactor fuel rods and container underwent damage between March 12 and 15, and a large amount of radioactive nuclides was emitted into the environment. As of April 12, it was confirmed that the cores for the No. 1 to No. 3 reactors were damaged and that the fuel pellet had melted. In light of this, the Fukushima Dai-ichi nuclear power plant accident was elevated to a level 7 event on the International Nuclear Event Scale (INES) (Releases, 2011). The radioactive contamination in the environment is become a great concern in Japan for our survival.

In this study, environmental samples of soil, plant species, and water were collected on April 10—about one month after the accident—in front of the main gate of the Fukushima Dai-ichi nuclear power plant and 35 km away at Iitate village in Fukushima prefecture. Doses of radioactive nuclides were then measured by a Ge(Li) semiconductor detector without any chemical modification of the samples in order to comprehensively identify the nuclides that diffused into the environment.

2. Methods

Soil, plant species, and water in the environment were collected on April 10, 2011 in front of the main gate of the Fukushima Dai-ichi nuclear power plant (located at Okuma-machi, Futaba-gun) and at Iitate village, Soma-gun, Fukushima prefecture, shown in Table 1. From paddy and thicket soils, 30–80 g of soils from the surface (0–10 cm) was collected after removing plant species. From pine and straw leaves, *ca.* 10 g of plant were collected while ensuring the absence of attached soil. From paddy fields and puddles, we collected *ca.* 100 ml of water per sample using a 50-ml syringe. The skies were clear and there was hardly any wind on the sampling day. The sampling sites and spatial radiation dose at each site are shown in Fig. 4. The latter was measured by an ion-chamber-type gamma-ray detector and GM counter.

Gamma-rays from collected samples were measured using a Ge(Li) semiconductor detector (Princeton Gamma-Tec). The device, located at the Radioisotope Center in the University of Tokyo, has a relative efficiency of 34.4% at 1332 keV, and FWHM of 1.78 keV on the 1332 keV ^{60}Co γ-line and 743 eV on the 122 keV ^{57}Co γ-line. Measurements were performed on April 14 and April 22. The sample was sealed

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doi:10.1016/j.envpol.2012.01.001

Table 1
Sampling sites and spatial dose of radiation.

Sample			Place	Distance from the main gate (km)	Dose of radiation (μSv h^{-1})
s1	Soil	Paddy	Around the nuclear plant	1.5	75
s2		Paddy	Near Kumagawa river	4.1	43
s3		Garden plant	In front of the main gate	0.0	100
s4		Paddy	Nagadoro, Iitate village	35.0	17
s5		Paddy	Komiya, Iitate village	35.0	11
p1	Plant species	Straw	Around the nuclear plant	1.5	75
p2		Leaf of pine	In front of the main gate	0.0	100
p3		Leaf of pine	In front of the main gate	0.0	100
p4		Straw	Komiya, Iitate village	35.0	11
w5	Water	Puddle	Around the nuclear plant	1.0	67
w6		Paddy	Around the nuclear plant	1.5	75

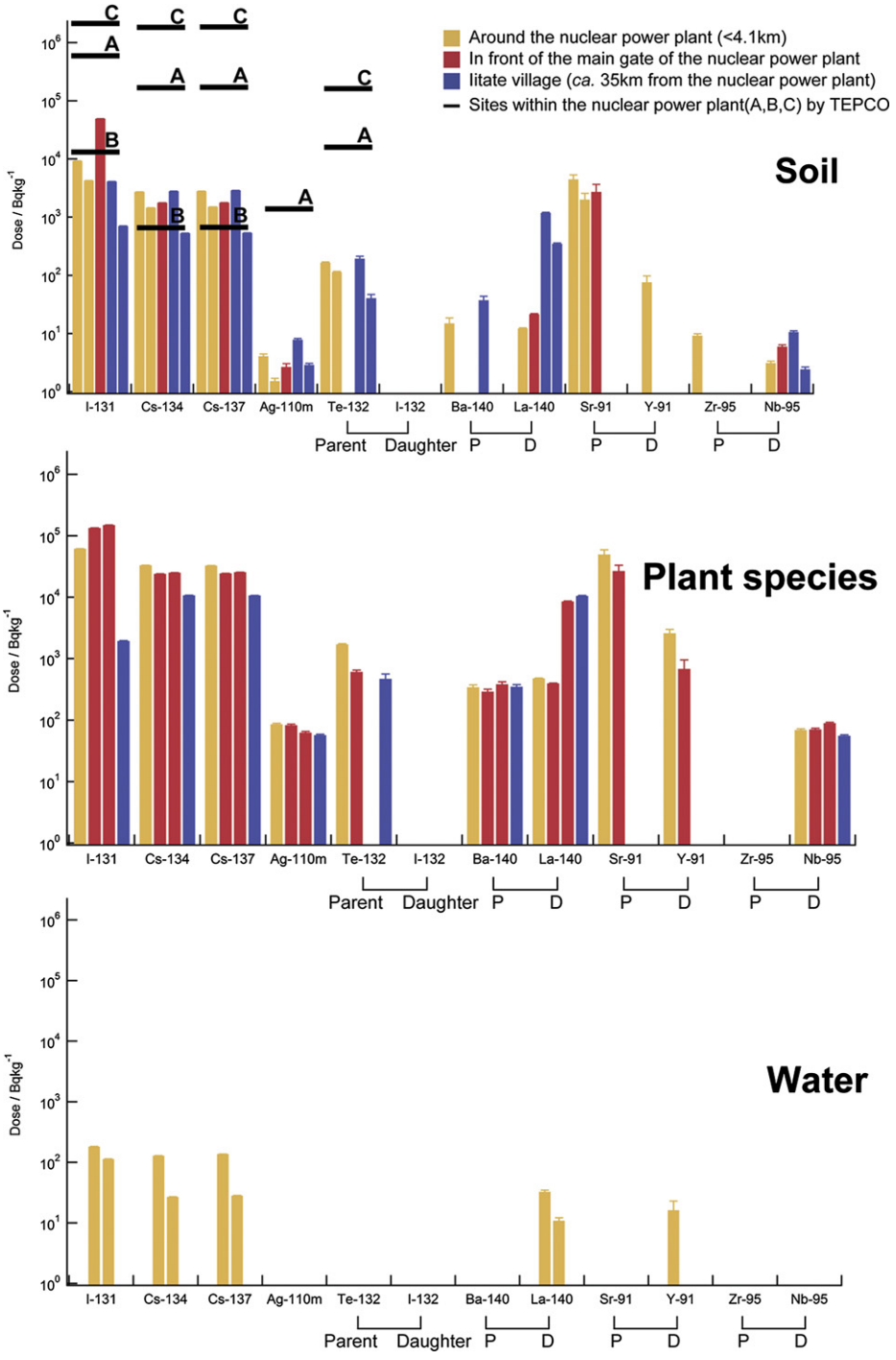

Fig. 1. Fission products (Bq kg^{-1}) in samples collected around the Fukushima Dai-ichi nuclear power plant and at Iitate village.

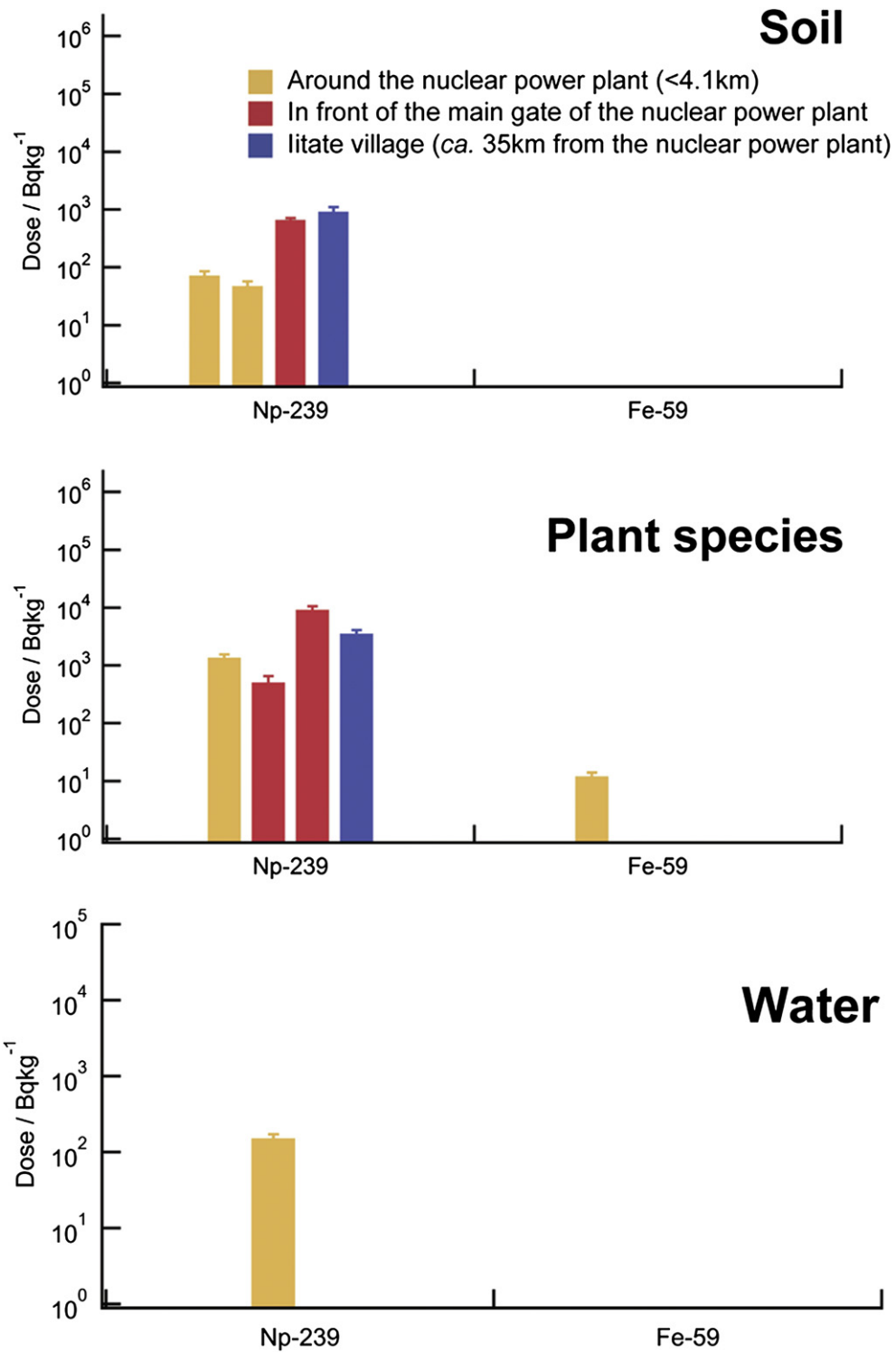

Fig. 2. Activation products (Bq kg^{-1}) in samples collected around the Fukushima Dai-ichi nuclear power plant and at Iitate village.

in a 100 ml vial made of polyethylene and subjected to measurement without chemical modification. The measurement time was 3000–3600 s. Spectrum navigator (SEIKO E&G Co., Ltd.) and a nuclide library generation program (NucLib ver 1.12) were used as the gamma-ray libraries for nuclide identification, together with the IAEA Handbook of Nuclear Data for Safeguards. A nuclide was identified when two or more clear and independent peaks greater than 3σ above baseline were present in the spectrum. The half-life correction of each nuclide was made assuming a sampling time of 0 s of the elapsed time.

The doses of identified nuclides in soil, plant species, and water samples collected in front of the power plant and at Iitate village are shown in Figs. 1 and 2. The fission products are listed in Fig. 1, and the activation products are listed in Fig. 2. The doses of nuclides in the surface soil collected on April 7 within the premises of the Fukushima Dai-ichi nuclear power plant, which were reported by Tokyo Electric Power Company; TEPCO (three fixed observation points within the plant; point A, playground; point B, in the woods; point C, disposal site), are also included in both Figures.

Radioactive elements, which were ^{131}I, ^{134}Cs (^{133}Cs), ^{137}Cs, ^{110m}Ag (^{109}Ag), ^{132}Te, ^{132}I, ^{140}Ba, ^{140}La, ^{91}Sr, ^{91}Y, ^{95}Zr, and ^{95}Nb were identified from each environmental sample. The nuclear data for these radionuclides are shown in Table 2. In the case of ^{132}I, which has a short half-life, its parent nuclide ^{132}Te was used to

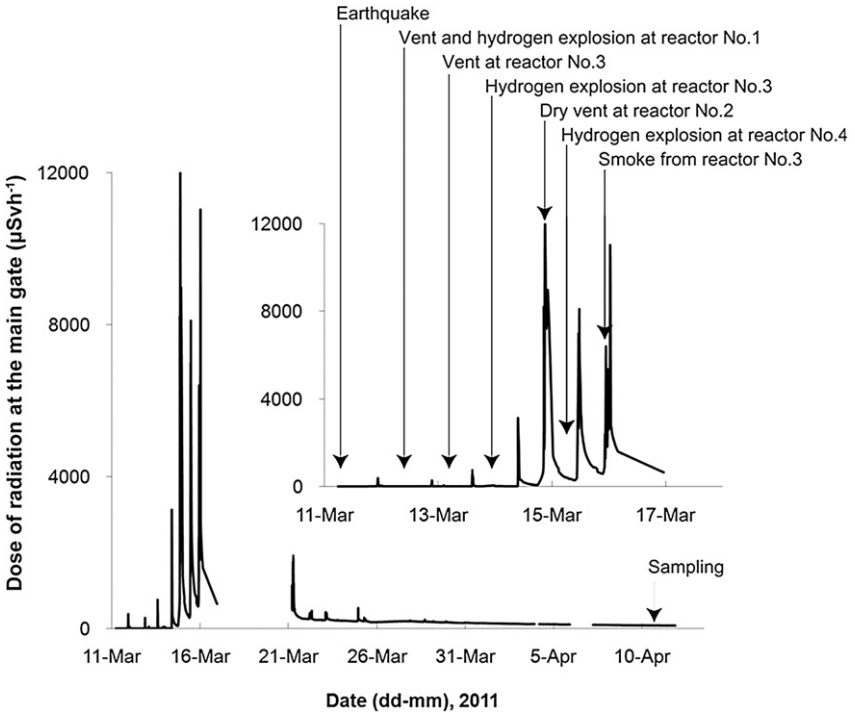

Fig. 3. Time series of the spatial dose of radiation at the main gate of the Fukushima Dai-ichi nuclear power plant.

estimate the half-life based on the radiation equilibrium. In the cases of ^{140}Ba–La, ^{91}Sr–Y, and ^{95}Zr–Nb, the doses for both nuclides are listed in Fig. 1 because the differences between them were small. For all nuclides, the doses in plant species were about 10 times higher than those in soil, which in turn were higher than those in water. In the case of ^{131}I, which is one of the main fission products, the dose was 49 kBq kg^{-1} in the soil collected in front of the main gate of the power plant, and 150 kBq kg^{-1} in plant species. On the other hand, the dose of ^{131}I in the soil at litate village (s4) was 4.1 kBq kg^{-1}, and of the same grade to that in the soil sample (s2) collected in a paddy field 4.1 km away from the nuclear power plant (4.3 kBq kg^{-1}). In the soil collected within the premises of the nuclear power plant site, the maximum observed dose was 2.3 MBq kg^{-1} reported by TEPCO on April 22.

These observations as of April 10 suggest that the dose of ^{131}I depended on the distance from the nuclear power plant. However, ^{134}Cs and ^{137}Cs, which have long half-lives, did not depend on the sampling point, as the dose ranges were 0.53–2.8 kBq kg^{-1} and 0.54–2.9 kBq kg^{-1}, respectively. ^{134}Cs is produced by neutron activation of ^{133}Cs, a stable fission product, whereas ^{137}Cs is produced directly from fission (Lin and Skarpelos, 1997). The ^{134}Cs/^{137}Cs ratio was *ca.* 1 at each sampling point. In particular, the highest dose was observed in a soil sample collected at Nagadoro in litate village. Based on a consideration of these measurements and half-life, we suggest that there was a very large but temporary diffusion of these radioactive nuclides. The same tendency is also observed for ^{110m}Ag. The production of ^{110m}Ag is confirmed by the ^{109}Ag (n,γ) to ^{110m}Ag reaction in the more mobile fission products of the Dragon gas-cooled reactor at Winfrith, UK (Flowers, 1986). After the Chernobyl accident, the dose of ^{110m}Ag in the soil was raised from 4.5 to 46.1 Bq kg^{-1} (average 14.3 Bq kg^{-1}) in Greece eight weeks (Papastefanou et al., 1988). Like typical fission products, this nuclide is characterized by fast diffusion.

Fig. 3 shows the spatial dose continuously observed in front of the nuclear power plant, as reported by TEPCO. Observations at the main gate were not conducted from March 17 to March 20. However, it is seen that the spatial dose rose dramatically several times before March 14. We assume that each nuclide spread during these events.

The activation products, ^{239}Np and ^{59}Fe were identified from each environmental sample. As in the fission products, the doses in plant species were high. Naturally, ^{239}Np does not normally exist because of its short half-life. ^{237}Np, which is a daughter nuclide of ^{241}Am, is distributed throughout the world in very slight amounts (Kelley et al., 1999). However, ^{239}Np is known to have spread at least 1500 km only two weeks after the Chernobyl accident (Henriksen and Saxebol, 1988).

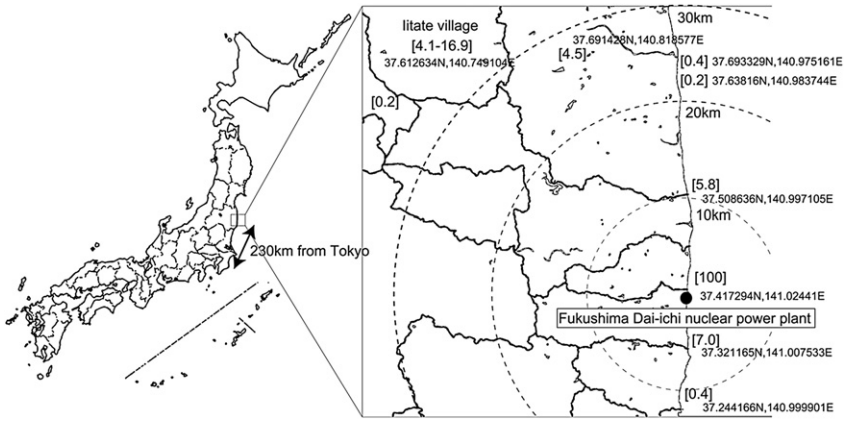

Fig. 4. Sampling sites and spatial dose of radiation. The value in square brackets, such as [100], is the value of the spatial dose of radiation (μSv h^{-1}) 1 m above ground on the day of sampling (April 10, 2011).

Table 2
Nuclear data for radionuclides by Evaluated Nuclear Data File, IAEA, in the environmental samples around Fukushima Dai-ichi nuclear power plant.

	Nuclide	Gamma (keV)	Abundance (%)	Half life (y: year) (d: day)(h: hour)
Fission	^{131}I	364.48	81.0	8.04(d)
	^{134}Cs(^{133}Cs)	604.66	97.6	2.06(y)
	^{137}Cs	661.64	85.0	30.2(y)
	^{110m}Ag(^{109}Ag)	884.67	72.8	252(d)
	^{132}Te	228.16	88.0	78.2(h)
	^{132}I	667.69	98.7	2.28(h)
	^{140}Ba	537.27	23.6	12.8(d)
	^{140}La	1596.49	95.5	40.3(h)
	^{91}Sr	555.57	61.0	9.48(h)
	^{91}Y	1208.00	0.3	58.5(d)
	^{95}Zr	756.72	54.6	64.0(d)
	^{95}Nb	765.79	99.8	35.0(d)
Activation	^{239}Np	106.14	27.8	2.35(d)
	^{59}Fe	1099.22	56.5	44.6(d)

^{239}Np was identified by each of the following peaks: 106.14 keV (27.8% of emission rate), 277.60 keV (14.5%), and 120.60 keV (2.77%) by Evaluated Nuclear Data File, IAEA, all of which show clear separation in the spectrum. The peaks at 117.26 and 228.20 keV were not used for identification because they are interfered by ^{132}Te. Given that ^{239}Np has a short half-life of 2.3 days, the gamma-ray spectrum was measured again nine days after the first measurement. By this time, all of the peaks decreased below the minimum limit of detection. ^{239}Np is a beta decay nuclide of ^{239}U, and ^{239}U is generated in large quantities by the neutron capture of ^{238}U in a nuclear reactor, as in the reaction $^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U}\xrightarrow{\beta}\mathrm{Np}\xrightarrow{\beta}{}^{239}\mathrm{Pu}$. Although it is unknown whether this nuclide spread between March 12 and March 15, it spread at least to Iitate village on April 10—the time of sample collection. In particular, a 3.6 kBq kg^{-1} dose of ^{239}Np was detected in the straw sample collected at the Komiya area in Iitate village. This dose was one third of that in the pine leaves in front of the main gate (9.2 kBq kg^{-1}). As the daughter nuclide of ^{239}Np is ^{239}Pu, diffusion of the former may cause serious contamination. From 1945 to 1980, 6500 TBq of ^{239}Pu was emitted in a global fallout, whereas the fallout from Chernobyl was 30 TBq (Beasley et al., 1998; Radiation, 2000). Although ^{239}Np emission from the Fukushima Dai-ichi nuclear power plant cannot be presumed at present, it is expected that ^{239}Pu will be detected at high doses around the power plant in the future. In addition, ^{59}Fe is the activation product from fuel cladding and impurities in structural materials (Thind, 2001). ^{59}Fe are presumed to be the activation products of ^{58}Fe obtained by the corrosion of cooling pipes. These nuclides spread into the environment by the venting operation or hydrogen explosion. ^{59}Fe, which is a typical neutron-induced nuclide, was also detected in the soil near the site of the critical accident at Tokai-mura, Japan, in 1999 (Nakanishi et al., 2000). This constitutes circumstantial evidence of ^{59}Fe being generated by neutron activation. At present, it is difficult to identify the nuclear reactor from which these nuclides were emitted. As MOX fuel is used in reactor No. 3, the quantification of Pu may yield information on this issue. However, as this is a time-consuming process, we do not address this here.

Acknowledgement

This work was carried out under the support of Radioisotope Center, The University of Tokyo.

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