Radiation Protection Dosimetry (2012), Vol. 151, No. 3, pp. 537–545 doi:10.1093/rpd/ncs040
Advance Access publication 13 April 2012

ENVIRONMENTAL CONTAMINATION AND EXTERNAL RADIATION DOSE RATES FROM RADIONUCLIDES RELEASED FROM THE FUKUSHIMA NUCLEAR POWER PLANT

Yasuyuki Taira[1,6], Naomi Hayashida[1], Shunichi Yamashita[2], Takashi Kudo[3], Naoki Matsuda[4], Jumpei Takahashi[5], Alexander Gutevitc[7], Alexander Kazlovsky[8] and Noboru Takamura[1,*]
[1]Department of Global Health, Medical, and Welfare, Nagasaki University Graduate School of Biomedical Sciences, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan
[2]Department of Radiation Medical Science, Nagasaki University Graduate School of Biomedical Sciences, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan
[3]Department of Radioisotope Medicine, Nagasaki University Graduate School of Biomedical Sciences, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan
[4]Division of Radiation Biology and Protection Center for Frontier Life Sciences, Nagasaki University Graduate School of Biomedical Sciences, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan
[5]Center for International Collaborative Research, Nagasaki University, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan
[6]Nagasaki Prefectural Institute for Environmental Research and Public Health, 2-1306-11 Ikeda Omura, Nagasaki 856-0026, Japan
[7]Zhitomir Inter-Area Medical Diagnostic Center, Kievskaya 21b, Korosten 260100, Ukraine
[8]Gomel State Medical University, 5 Lange Street, Gomel 240600, The Republic of Belarus

*Corresponding author: takamura@nagasaki-u.ac.jp

Received November 16 2011, revised March 5 2012, accepted March 5 2012

To evaluate the environmental contamination and contributory external exposure after the accident at the Fukushima Nuclear Power Plant (FNPP), the concentrations of artificial radionuclides in soil samples from each area were analysed by gamma spectrometry. Six artificial radionuclides (^{131}I, ^{134}Cs, ^{137}Cs, ^{129m}Te, ^{95}Nb and ^{136}Cs) were detected in soil samples around FNPP. Calculated external effective doses from artificial radionuclide contamination in soil samples around FNPP were 1.9–2.9 μSv h^{-1} (8.7–17.8 mSv y^{-1}) in Fukushima city on 22 March 2011. After several months, these calculated external effective doses were 0.25–0.88 μSv h^{-1} (2.2–7.6 mSv y^{-1}) in Fukushima city on 29 June 2011. The present study revealed that the detected artificial radionuclides around FNPP mainly shifted to long-lived radionuclides such as radioactive caesium (^{134}Cs and ^{137}Cs) even though current levels are decreasing gradually due to the decay of short-lived radionuclides such as ^{131}I, ^{129m}Te, ^{95}Nb and ^{136}Cs. Thus, radiation exposure potency still exists even though the national efforts are ongoing for reducing the annual exposure dose closer to 1 mSv, the public dose limit. Long-term environmental monitoring around FNPP contributes to radiation safety, with a reduction in unnecessary exposure to the residents.

INTRODUCTION

On 11 March 2011, a 9.0-magnitude earthquake (The Great East Japan Earthquake) struck the east coast near Iwate, Miyagi and Fukushima, Japan. The earthquake, in combination with the tsunami, caused extensive damage to the Fukushima Dai-ichi Nuclear Power Plant (FNPP), and convergence of the accident is not completely settled although recovery efforts are ongoing. Recently, it is estimated that various radioactive elements have been released by hydrogen detonation and damage to the reactors since March 12. The radioactive plume derived from Units 1, 2, 3 and 4 of FNPP has dispersed in the atmosphere. The Nuclear Safety Commission of Japan estimated and announced the result of the trial calculation in the current stage regarding the total amount of radioactive materials discharged into the atmosphere from the results of monitoring the data of ^{131}I and ^{137}Cs as the total amount of the discharge from FNPP. The resulting value corresponds to 'Level 7' of the international nuclear and radiological event scale (INES) by the International Atomic Energy Agency (IAEA). Although this level corresponds to a major accident, it is estimated that the amount of radioactive materials discharged into the environment in the current stage is ~10 % (1.6×10^{17} Bq for ^{131}I and 1.5×10^{16} Bq for ^{137}Cs) of the accident at the Chernobyl Nuclear Power Plant (CNPP), which was previously assessed at the same level (http://www.kantei.go.jp/foreign/kan/topics/201106/iaea_houkokusho_e.html).

The two main pathways leading to radiation exposure of the general public due to 'fallout' are external exposure from radionuclides deposited on the

ground and internal exposure through the ingestion of contaminated foods produced in contaminated areas. It is extremely important to evaluate the environmental contamination as well as the external and internal exposure risks due to nuclear disasters for radiation protection and public health. Risks of internal exposure are extremely low because restrictions of food intake by the nation are strictly carrying out after the FNPP accident (http://www.mhlw.go.jp/english/topics/2011eq/index.html). On the other hand, risks of external exposure around living space are becoming a matter of public attention to confirm the safety to lives. Therefore, for the evaluation of current environmental contamination and contributions from external exposure due to artificial radionuclides, concentrations of radionuclides in soil samples from resident areas around FNPP were analysed by gamma spectrometry. Furthermore, external effective doses were calculated from samples from these areas for the estimation of radiation exposure status.

MATERIALS AND METHODS

Sampling places

FNPP is located on the east coast of the island of Honshu, ~200 km northeast of Tokyo. Samples around FNPP were collected from: Fukushima city (the prefectural capital, 37°41′N, 140°28′E), 57.8 km northwest of FNPP (37°25′N, 141°02′E); Iwaki city (37°03′N, 140°53′E), 43.4 km south of FNPP; Ono town (37°17′N, 140°37′E), 39.1 km southwest of FNPP and Iitate village (37°40′N, 140°44′E), 39.1 km northwest of FNPP in Japan (Figure 1).

Measurement of radionuclides

For the evaluation of external radiation exposure, undisturbed surface soils were collected by a core sampling technique (12 samples at soil depths of 0–5 and 5–10 cm) around FNPP between March and July 2011 (Fukushima city on 22 March and 29 June Iwaki city on 7April, Ono town on 7 April and Iitate village on 11 July, respectively). Six samples were collected at Fukushima Medical University (Fukushima city), and two samples were collected at Iwaki common building (Iwaki city), Ono town office (Ono town) and Iitate village office (Iitate village), respectively. The quantity of soil collected in each area was between 8.8 and 106 g. After collection, all samples were dried by a fixed temperature dryer (105°C, 24 h) and sieved for pebbles and organic materials (<2 mm) before measurement of radionuclide activity.

After preparation, samples were put in plastic containers made of polypropylene and analysed with a high-purity germanium detector (ORTEC®, GEM35, Ortec International Inc., Oak Ridge, TN, USA) coupled to a multichannel analyzer

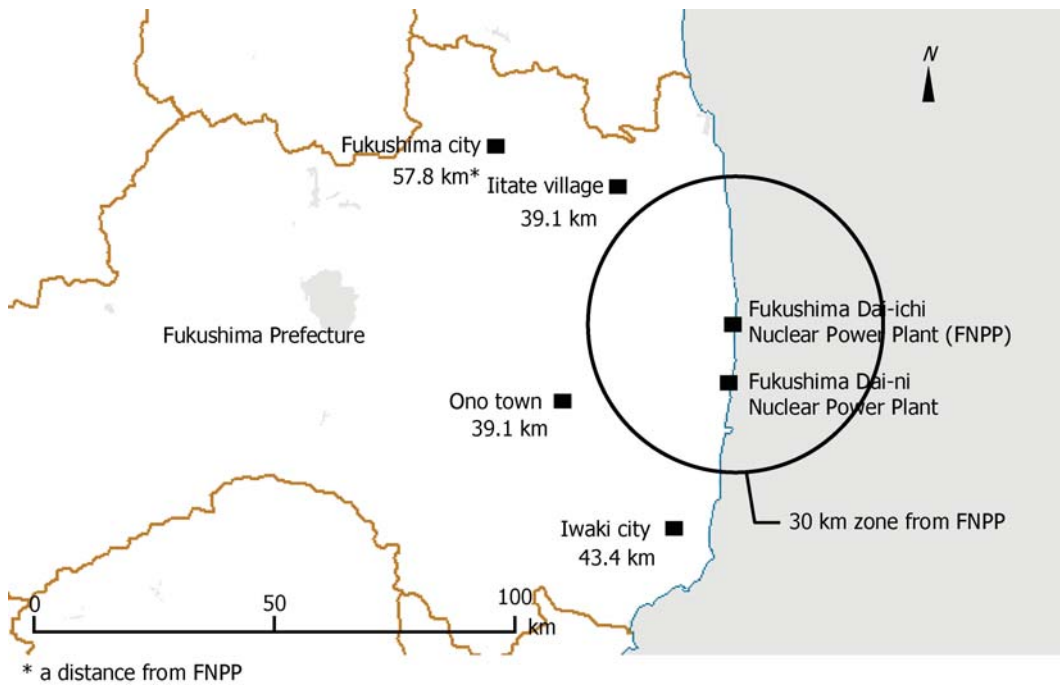

Figure 1. Areas around the FNPP.

(MCA7600, Seiko EG &G Co., Ltd., Chiba, Japan) for 7200 s (FNPP). The authors set the measuring time for that at which objective radionuclides could be detected sufficiently, although they must reinforce the monitoring of the environmental after the FNPP accident. Gamma-ray peaks used for measurements were 364.48 keV for ^{131}I, 604.66 keV for ^{134}Cs, 661.64 keV for ^{137}Cs, 695.98 keV for ^{129m}Te, 765.79 keV for ^{95}Nb and 818.50 keV for ^{136}Cs. Decay corrections were made based on sampling date. Detector efficiency calibration for different measurement geometries was performed using mixed activity standard volume sources (Japan Radioisotope Association, Tokyo, Japan). The relative detection efficiency of this instrument was 37.7 %. Sample collection, processing and analysis were executed in accordance with standard methods of radioactivity measurement authorised by the Ministry of Education, Culture, Sports, Science and Technology, Japan (http://www.kankyo-hoshano.go.jp/en/index.html)[1]. All measurements were performed at Nagasaki Prefectural Institute for Environmental Research and Public Health (Nagasaki, Japan).

Effective dose

After measurements, external effective doses from soil samples were calculated from artificial radionuclide concentrations with the following formula:

$$H_{\text{ext}} = C \cdot D_{\text{ext}} \cdot f \cdot s$$

in which C is the activity concentration of detected artificial radionuclides (kBq m^{-2}; calculated from radionuclide concentration in Bq kg^{-1} and collected areas of surface soil (0–5 cm)); D_{ext} is the dose conversion coefficient reported as the kerma-rate in air at 1 m above ground per unit activity per unit area ((μGy h^{-1})/(kBq m^{-2})) for detected artificial radionuclides with the relaxation mass per unit area (β, g cm^{-2}) set to 1.0 due to the passage of 3–6 weeks immediately after the accident at FNPP and set to 2.0 due to the passage of <1 y after the accident at FNPP (dose conversion coefficients are shown in the Appendix)[2]; f is the unit conversion coefficient (0.7 Sv Gy^{-1} for effective dose rate in the body per unit absorbed dose rate in air)[3]; s is the occupancy-shielding factor (0.2 fractional time outdoors+0.8 fractional time indoors×0.2 building shielding= 0.36)[3]. Also, annual external effective doses due to radioactive caesium (^{134}Cs and ^{137}Cs) were calculated using Equation (1) (H_{ext}×24 h×365 d).

Furthermore, external effective doses of monitoring personnel who sampled soil around FNPP were estimated from a portable detector for the management of radiation exposure (PDM-112®, Hitachi-Aloka Medical, Ltd., Tokyo, Japan).

RESULTS

The distribution of detected artificial radionuclides in surface soil samples around FNPP are shown in Table 1. Immediately after the accident, the prevalent dose-forming artificial radionuclides from soil samples around FNPP were ^{131}I, ^{134}Cs, ^{137}Cs, ^{129m}Te, ^{95}Nb and ^{136}Cs (these concentrations are shown in Table 1). In several months after the accident, the prevalent dose-forming artificial radionuclides from soil samples around FNPP were ^{134}Cs, ^{137}Cs and ^{129m}Te (these concentrations are shown in Table 1).

Next, the vertical distribution of detected artificial radionuclides in soil samples around FNPP are shown in Table 2. The concentrations of detected artificial radionuclides in surface soils around FNPP were higher than those of lower layers and these radionuclides were mainly accumulated in surfaces (these concentrations are shown in Table 2).

For estimating the external effective doses, the activity concentrations (in kBq m^{-2}) of detected artificial radionuclides in surface soils (0–5 cm) around FNPP were calculated from these radionuclide concentrations in Bq kg^{-1} (these concentrations are shown in Table 3).

The external effective doses from detected artificial radionuclides around FNPP using Equation (1) are summarised in Table 4. Immediately after the accident, calculated external effective doses around FNPP were 1.9–2.9 μSv h^{-1} (8.7–17.8 mSv y^{-1}) in Fukushima city, 0.058 μSv h^{-1} (0.092 mSv y^{-1}) in Iwaki city and 0.092 μSv h^{-1} (0.62 mSv y^{-1}) in Ono town, respectively. Several months after the accident, the calculated external effective doses around FNPP were 0.25–0.88 μSv h^{-1} (2.2–7.6 mSv y^{-1}) in Fukushima city and 0.53 μSv h^{-1} (4.6 mSv y^{-1}) in Iitate village, respectively (these doses are shown in Table 4).

Furthermore, the estimated doses from a portable detector for the management of monitoring personnel were 0.39–0.41 μSv h^{-1} around 30 km over the distance from the reactor (Fukushima city, Iwaki city and Ono town) from 29 March to 8 April 2011 and 0.17 μSv h^{-1} around 30 km over the distance from the reactor (Fukushima city, Soma city, Iwaki city, Ono town and Iitate village) from 5 to 12 July 2011.

DISCUSSION

Since 12 March 2011, a 20-km radius from FNPP has been stipulated as 'a no-entry zone'. In the emergency zone outside a 20-km radius from FNPP, there are certain areas where radioactive materials emitted from the power station have accumulated as a result of climatic and geographical conditions and in some localised areas, these accumulated volumes of radioactive materials are at high levels. Thus,

Table 1. Distribution (Bq kg^{-1}dry^{-1}) of detected artificial radionuclides in surface soils (0–5 cm) collected at areas around FNPP.

	Radionuclide concentration in Bq kg^{-1} dry^{-1}					
	^{131}I (8.0 d)[a]	^{134}Cs (2.1 y)	^{137}Cs (30 y)	^{129m}Te (34 d)	^{95}Nb (35 d)	^{136}Cs (13 d)
Fukushima city A (March)	54 150 ± 780[b] (1410)[c]	13 402 ± 85.6 (71)	12 989 ± 81.9 (51)	14 908 ± 1017 (2390)	n.d.[d] (78)	1649 ± 106 (241)
Fukushima city B (March)	34 287 ± 670 (1545)	20 838 ± 93.1 (77)	20 427 ± 89.5 (57)	20 288 ± 1137 (2797)	n.d. (82)	2521 ± 116 (258)
Fukushima city C (March)	18 775 ± 429 (991)	11 417 ± 59.8 (51)	11 158 ± 57.1 (36)	10 654 ± 721 (1800)	75.5 ± 18 (51)	1319 ± 74 (171)
Fukushima city D (June)	n.d. (313)	26 560 ± 70.2 (57)	28 690 ± 73.5 (43)	2531 ± 449 (1312)	n.d. (40)	n.d. (72)
Fukushima city E (June)	n.d. (129)	6107 ± 27.5 (24)	6648 ± 29.0 (18)	n.d. (582)	n.d. (18)	n.d. (30)
Fukushima city F (June)	n.d. (162)	12 836 ± 32.5 (27)	14 257 ± 34.9 (21)	1712 ± 254 (738)	n.d. (22)	n.d. (41)
Iwaki city (April)	3560 ± 33.2 (23)	146 ± 4.2 (6.2)	152 ± 4.4 (5.4)	631 ± 73 (169)	n.d. (6.6)	11.4 ± 2.7 (7.7)
Ono town (April)	1403 ± 26.2 (44)	1052 ± 10.6 (9.6)	1099 ± 11.1 (7.8)	672 ± 98 (264)	n.d. (7.7)	60.2 ± 5.7 (13)
Iitate village (July)	n.d. (77)	12 799 ± 41.8 (34)	14 151 ± 45.3 (28)	1629 ± 251 (725)	n.d. (21)	n.d. (25)

Samples were collected at areas around FNPP between March and July, 2011 (Fukushima city on 22 March and 29 June, Iwaki city on 7 April, Ono town on 7 April and Iitate village on 11 July). Radionuclides were analysed with a germanium detector coupled to a multichannel analyser for 7200 s at Nagasaki Prefectural Institute for Environmental Research and Public Health, Nagasaki, Japan.

[a]Half-life.

[b]Error shows one sigma standard deviation from counting statistics.

[c]Detection limit.

[d]Not detected.

Table 2. Vertical distribution of detected artificial radionuclides in soils (0–5 and 5–10 cm) collected at areas around FNPP.

	Radionuclide concentration in Bq kg^{-1} dry^{-1}					
	^{131}I (8.0 d)[a]	^{134}Cs (2.1 y)	^{137}Cs (30 y)	^{129m}Te (34 d)	^{95}Nb (35 d)	^{136}Cs (13 d)
Iwaki city (0–5 cm) (April)	3560 ± 33.2[b] (23)[c]	146 ± 4.2 (6.2)	152 ± 4.4 (5.4)	631 ± 73 (169)	n.d.[d] (6.6)	11.4 ± 2.7 (7.7)
Iwaki city (5–10 cm) (April)	2219 ± 30.8 (26)	138 ± 4.3 (5.2)	148 ± 4.5 (4.6)	310 ± 69 (194)	n.d. (7.0)	n.d. (16)
Ono town (0–5 cm) (April)	1403 ± 26.2 (44)	1052 ± 10.6 (9.6)	1099 ± 11.1 (7.8)	672 ± 98 (264)	n.d. (7.7)	60.2 ± 5.7 (13)
Ono town (5–10 cm) (April)	325 ±14 (26)	217 ± 5.1 (5.0)	223 ± 5.4 (5.3)	n.d. (172)	n.d. (5.1)	n.d. (16)
Iitate village (0–5 cm) (July)	n.d. (77)	12 799 ± 41.8 (34)	14 151 ± 45.3 (28)	1629 ± 251 (725)	n.d. (21)	n.d. (25)
Iitate village (5–10 cm) (July)	n.d. (58)	5256 ± 28.7 (24)	5788 ± 30.7 (20)	n.d. (481)	n.d. (15)	n.d. (19)

Samples were collected at areas around FNPP between April and July 2011 (Iwaki city on 7April, Ono town on 7 April and Iitate village on 11 July). Radionuclides were analysed with a germanium detector coupled to a multichannel analyser for 7200 s at Nagasaki Prefectural Institute for Environmental Research and Public Health, Nagasaki, Japan.
[a]Half-life.
[b]Error shows one sigma standard deviation from counting statistics.
[c]Detection limit.
[d]Not detected.

these emergency zones are extremely important areas for practising environmental monitoring until the evacuation zones are re-designated.

The present study confirmed that the areas located northwest of FNPP, including 'planned evacuation zones' (http://www.kantei.go.jp/foreign/incident/index.html) such as Iitate village over 30 km from FNPP, are obviously affected by the accident compared with other regions in Fukushima prefecture because at most six artificial radionuclides, including short-lived radionuclides such as ^{131}I, ^{129m}Te, ^{95}Nb and ^{136}Cs, were detected in the soil in Fukushima and confirmed that detected artificial radionuclides in soils were mainly distributed in the depth of 0–5 cm (Tables 1 and 2). Radionuclides are generally transported by adhesion to aerosol or soil particles, and weather condition such as rain, snow and wind is likely to play a crucial role in transporting radionuclides from FNPP. It is suggested that a part amount of radionuclide deposits on the surface soil and others are transported to underground with rain or are transferred to some other places by re-floating. Radionuclide analysis of environmental samples is extremely practical for the evaluation of current environmental radioactivity levels.

According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), areas of ^{137}Cs deposition density greater than 555 kBq m^{-2} (15 Ci km^{-2}) are designated as areas of strict control about the CNPP accident on 29 April 1986[3]. Long-term (10 d) and complicated dynamics of the release of radioactive substances from Unit 4 of CNPP during the accident and changes in the meteorological conditions during this period have resulted in a composite picture of contamination of vast territories[4]. Various radioactive elements such as radioactive noble gases, iodine isotopes, radio-caesium and less-volatile radionuclides were released from the reactor into the atmosphere[5]. During the first weeks after the CNPP accident, most of the activity deposited on the ground consisted of short-lived radionuclides such as ^{131}I[3]. Deposition in the nearby contaminated zone (<100 km) reflected the radionuclide composition of the fuel, and the volatile elements, including iodine and caesium, in the form of condensation-generated particles were more widely dispersed into the far zone (from 100 km to ~2000 km)[3]. The ^{137}Cs deposition was highest within a 30-km radius area surrounding the reactor, known as the 30-km zone, and deposition densities exceeded 1500 kBq m^{-2} in this zone and some areas (Gomel, Kiev and Zhitomir regions) of the near zone to the west and northwest of the reactor[3]. Nearly, 400 million people resided in territories that were contaminated with radioactivity at a level higher than 4 kBq m^{-2} (0.11 Ci km^{-2}) from April to July 1986[6]. In 2000, the total inventories of the fuel component

Table 3. Distribution (kBq m^{-2}) of detected artificial radionuclides in surface soils (0–5 cm) collected at areas around FNPP.

	Radionuclide concentration in kBq m^{-2}					
	^{131}I (8.0 d)[a]	^{134}Cs (2.1 y)	^{137}Cs (30 y)	^{129m}Te (34 d)	^{95}Nb (35 d)	^{136}Cs (13 d)
Fukushima city A (March)	2615 ± 37.6[b]	647 ± 4.1	627 ± 4.0	720 ± 49.1	n.d.[c]	79.6 ± 5.1
Fukushima city B (March)	2160 ± 42.2	1313 ± 5.9	1287 ± 5.6	1278 ± 71.7	n.d.	159 ± 7.3
Fukushima city C (March)	1553 ± 35.5	944 ± 4.9	923 ± 4.7	881 ± 59.6	6.2 ± 1.4	109 ± 6.1
Fukushima city D (June)	n.d.	489 ± 1.3	528 ± 1.4	46.6 ± 8.3	n.d.	n.d.
Fukushima city E (June)	n.d.	185 ± 0.8	201 ± 0.9	n.d.	n.d.	n.d.
Fukushima city F (June)	n.d.	651 ± 1.6	724 ± 1.8	86.9 ± 13	n.d.	n.d.
Iwaki city (April)	163.0 ± 1.5	6.7 ± 0.2	7.0 ± 0.2	28.9 ± 3.3	n.d.	0.5 ± 0.1
Ono town (April)	59.9 ± 1.1	45.0 ± 0.5	47 ± 0.5	28.7 ± 4.2	n.d.	2.6 ± 0.2
Iitate village (July)	n.d.	395 ± 1.3	436 ± 1.4	50.2 ± 7.7	n.d.	n.d.

[a]Half-life.
[b]Error shows one sigma standard deviation from counting statistics.
[c]Not detected.

Table 4. External effective doses from artificial radionuclides in areas around FNPP.

	External effective dose		Mainly contributed radionuclides
	μSv h^{-1}[a]	mSv y^{-1}[b]	
Fukushima city A (March)	1.9	8.7	^{131}I, ^{134}Cs
Fukushima city B (March)	2.9	17.8	^{131}I, ^{134}Cs
Fukushima city C (March)	2.1	12.8	^{131}I, ^{134}Cs
Fukushima city D (June)	0.65	5.7	^{134}Cs, ^{137}Cs
Fukushima city E (June)	0.25	2.2	^{134}Cs, ^{137}Cs
Fukushima city F (June)	0.88	7.6	^{134}Cs, ^{137}Cs
Iwaki city (April)	0.058	0.092	^{131}I, ^{134}Cs
Ono town (April)	0.092	0.62	^{131}I, ^{134}Cs, ^{137}Cs
Iitate village (July)	0.53	4.6	^{134}Cs, ^{137}Cs
Over 30 km distance from FNPP reactor (March–April)	0.39–0.41[c]		
Over 30 km distance from FNPP reactor (July)	0.17		

[a]External effective doses were calculated with the following formula: $H_{ext} = C \cdot D_{ext} \cdot f \cdot s$, where C is the activity concentration of detected artificial radionuclides (kBq m^{-2}; calculated from radionuclide concentration in Bq kg^{-1} and collected areas of soils (0–5 cm)), D_{ext} is the dose conversion coefficient as kerma-rate in air at 1 m above ground per unit activity per unit area ((μGy h^{-1})/(kBq m^{-2}) for detected artificial radionuclides with the value of relaxation mass per unit area 1.0 and/or 2.0 g cm^{-2}[2]), f is the unit conversion coefficient (0.7 Sv Gy^{-1}[3]), s is the occupancy-shielding factor (0.2 fractional time outdoors+0.8 fractional time indoors×0.2 building shielding=0.36[3]).
[b]Calculated external effective doses due to radioactive caesium (^{134}Cs and ^{137}Cs).
[c]External effective doses were estimated from a portable detector of monitoring personnel who sampled soils around FNPP (PDM-112®, Hitachi-Aloka Medical, Ltd).

radionuclides in the upper 30 cm of the soil layer in the 30-km Chernobyl zone in Ukraine were estimated as 0.4–0.5 % of the radionuclide amounts in the CNPP Unit 4 at the moment of the accident[6]. According to the 2006 IAEA report, the external doses around CNPP during 1986–2005 were ~1.2 times higher, and internal doses were ~1.1–1.5 times higher, than those obtained during 1986–1995 (depending on soil properties and applied countermeasures)[7]. Moreover, a previous study by the present authors revealed that areas around CNPP, especially the Russian–Belarussian border including Klincy city and Gomel oblast, are still contaminated[8]. Although the amounts of artificial radionuclides released from nuclear reactors and diffusion scales differ between FNPP and CNPP, the data of

the environmental radioactivity level around CNPP are extremely important for taking countermeasures against radiation exposure in the future of Fukushima.

It is also clear that external effective doses from detected artificial radionuclides around FNPP immediately after the accident were predominantly the comparatively short-lived radionuclides ^{131}I and ^{134}Cs. At several months after the accident, detected artificial radionuclides around FNPP mainly shifted to radioactive caesium (^{134}Cs and ^{137}Cs, Table 1). In general, the volatile elements primarily consist of radioactive iodine, including ^{131}I, which has a radioactive half-life of 8 d, and radioactive caesium, including ^{137}Cs with a half-life of 30 y. These volatile elements constitute aerosols and finally fall on the ground. The short-lived radionuclides should decay after a few months and should not therefore remain on any large-scale for a long time. Actually, 3 months have passed since the Fukushima accident and the shorter-lived radionuclides such as ^{131}I are decaying gradually (http://www.mext.go.jp/english/). In the CNPP accident, more than 4000 cases of thyroid cancer were diagnosed during 1986–2002 among those who were children or adolescents (0–17 y) at the time of the Chernobyl accident in Belarus, in Ukraine and in the four most contaminated regions of Russia[9]. Most of the thyroid dose was caused by the intake of ^{131}I with food during the first weeks after the Chernobyl accident. The shorter-lived radioiodines decayed quickly during the food-chain transport and their contribution is estimated to have been on the order of 1 % of the thyroid dose due to ^{131}I[10, 11]. The collective effective dose received during 1986–2005 by ~5 million residents living in affected areas of Belarus, Ukraine and Russia was ~50 000 man Sv with ~40 % from ingestion[7]. That contribution might have been larger if countermeasures had not been applied[7]. In the present study, current internal exposure through the consumption of locally produced food around FNPP may be very small for countermeasures against the health effects on the human body. However, estimated external effective doses due to radioactive caesium of Fukushima city and Iitate village located in the northwest of FNPP exceeded 1 mSv y^{-1}, the public dose limit[12] (Table 4). The national governments aims to responsibly promote the decontamination in areas with emergency exposure situations (i.e. annual exposure does is >20 mSv) and to reduce the annual exposure does to 1 mSv^{-1} in areas with existing exposure situations (areas where the annual exposure does is <20 mSv), in cooperative with municipalities and local residents through the implementations of effective decontamination work[13]. Especially, the government aims to reduce their estimated annual exposure dose closer to 1 mSv as early as possible and continue with further reductions in children's living spaces (such as schools or parks). Current situation after the FNPP accident corresponds to a radiological emergency and post-accident rehabilitation. Thus, it is necessary to maintain objective correspondence and detailed monitoring of the environmental radioactivity. Also, daily monitoring of the environmental radioactivity level around resident areas becomes the fundamental data for health effects by chronic and a low-level exposure such as radioactive caesium, especially ^{137}Cs.

There were several limitations in this study. Several radionuclides could not be analysed by an extraction procedure, including ^{90}Sr, and homogenised soil samples in extensive areas could not be standardised. Radionuclides in soil samples may be unequally distributed around FNPP because the size of the sample collection was small under the serious and emergent conditions of the FNPP accident. Although artificial radionuclides such as ^{131}I, ^{134}Cs and ^{137}Cs were detected at high levels in the areas around FNPP immediately after the accident, these radionuclides could be analysed sufficiently at very small quantities. However, further investigation with detailed conditions is needed. It is extremely important to estimate effective doses from the environmental samples such as soil by gamma spectrometry. The soil contamination due to the dispersion of fallout derived from the accident becomes a serious problem in Fukushima. Residents, especially children, adolescents and pregnant women, are anxious about the adverse health effects of radiation exposure from the environment including soil, plants and tap water. Moreover, residents who are engaged in farming or fishery worry about this circumstance because artificial radionuclides in foodstuffs such as crops and fishes are transferred to humans in the food chain. Since 1991, a change in this development has been observed: many measurements show stagnation or in some cases, even an increase in the contamination of foodstuff and humans in the Bryansk-Gomel Spot, although only a few groups of foodstuffs, such as potatoes, show a slight decrease in radioactivity[14]. However, it was reported that reduced plant uptake of radionuclides, especially ^{137}Cs and ^{90}Sr, by competitive cations contained in chemical fertilisers has a general advantage in large-scale, low-level contamination incidents on arable land and has been widely practised in central and western Europe after the Chernobyl accident[15]. The existing remediation approaches and phytoextraction (phytoremediation) of radionuclides from contaminated soils have been examined[16]. Thus, the remediation of contaminated soil by artificial radionuclides is a crucial social responsibility in Japan and internationally.

In conclusion, the external exposure potencies derived from the FNPP accident were evaluated.

The present study revealed that artificial radionuclides derived from the accident were detected from soil samples in areas around FNPP, even though current levels are decreasing gradually for the decay of short-lived radionuclides. Furthermore, dose rates from external exposure around FNPP immediately after the accident, especially in Fukushima city, were high compared with other areas around FNPP and contributed to the comparatively short-lived radionuclides such as ^{131}I and ^{134}Cs. Several months after the accident, the detected artificial radionuclides around FNPP mainly shifted to radioactive caesium (^{134}Cs and ^{137}Cs). Thus, radiation exposure potency still exists, even though the national efforts are ongoing for reducing the annual exposure dose closer to 1 mSv, the public dose limit (ICRP, 1991). Even though current internal exposure through the consumption of locally produced food around FNPP may be very small for countermeasures against the health effects on the human body, a long-term follow-up of environmental monitoring around FNPP, as well as evaluation of the health effects in the population residing around these areas, could contribute to radiation safety and reduce unnecessary exposure to residents.

ACKNOWLEDGEMENTS

The authors thank Mr. Shinichi Nakamura and Mr. Kohji Yoshida for assistance with environmental sampling and monitoring around the Fukushima Nuclear Power Plant.

FUNDING

This work was supported by the Ministry of Education, Culture, Sports, Science and Technology of Japan through the Nagasaki University Global COE program and by the Japan Society for the Promotion of Science Grant-in-Aid for Scientific Research 23933016.

REFERENCES

1. Ministry of Education, Culture, Sports, Science, and Technology, Japan. *Environmental radioactivity and radiation in Japan*. http://www.kankyo-hoshano.go.jp/en/index.html. (last accessed 25 February 2012).
2. International Commission on Radiation Units and Measurements. *Gamma-ray spectrometry in the environment.* ICRU Report 53. International Commission on Radiation Units and Measurements (1994).
3. United Nations Scientific Committee on the Effects of Atomic Radiation. *Sources and effects of ionizing radiation. 2000 Report to General Assembly, with scientific annexes.* United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations (2000).
4. Kulan, A. *Seasonal ^{7}Be and ^{137}Cs activities in surface air before and after the Chernobyl event*. J. Environ. Radioact. **90**, 140–150 (2006).
5. Kashparov, V. A., Lundin, S. M., Zvarych, S. I., Yoshchenko, V. I., Levchuk, S. E., Khomutinin, Y. V., Maloshtan, I. M. and Protsak, V. P. *Territory contamination with the radionuclides representing the fuel component of Chernobyl fallout*. Sci. Total Environ. **317**, 105–119 (2003).
6. Yablokov, A. V. and Nesterenko, V. B. *Chernobyl contamination through time and space.* Ann. N Y Acad. Sci. **1181**, 5–30 (2009).
7. Balonov, M. I., Anspaugh, L. R., Bouville, A. and Likhtarev, I. A. *Contribution of internal exposures to the radiological consequences of the Chernobyl accident*. Radiat. Prot. Dosim. **127**, 491–496 (2007).
8. Taira, Y., Hayashida, N., Brahmanandhan, G. M., Nagayama, Y., Yamashita, S., Takahashi, J., Gutevitc, A., Kazlovsky, A., Urazalin, M. and Takamura, N. *Current concentration of artificial radionuclides and estimated radiation doses from ^{137}Cs around the Chernobyl nuclear power plant, the Semipalatinsk nuclear testing site, and in Nagasaki*. J. Radiat. Res. **52**, 88–95 (2011).
9. World Health Organization. *Health effects of the Chernobyl accident and special health care programmes.* In: Report of the UN Chernobyl Forum Expert Group 'Health', Bennet, B., Repacholi, M. and Zhanat, C., Eds. World Health Organization, pp. 23–53 (2006).
10. Balonov, M., Kaidanovsky, G., Zvonova, I., Kovtun, A., Bouville, A., Luckyanov, N. and Voillequé, P. *Contributions of short-lived radioiodines to thyroid doses received by evacuees from the Chernobyl area estimated using early in-vivo activity measurements.* Radiat. Prot. Dosim. **105**, 593–599 (2003).
11. Gavrilin, Y. *et al. Individual thyroid dose estimation for a case-control study of Chernobyl-related thyroid cancer among children of Belarus. Part 1: 131I, short-lived radioiodines (132I, 133I, 135I), and short-lived radiotelluriums (131mTe and 132Te)*. Health Phys. **86**, 565–585 (2004).
12. International Commission on Radiological Protection. *The 1990 Recommendations of the International Commission on Radiological Protection.* ICRP Publication 60. Ann. ICRP **21**(1–3). International Commission on Radiological Protection (1991).
13. International Commission on Radiological Protection. *The 2007 Recommendations of the International Commission on Radiological Protection.* ICRP Publication 103. Ann. ICRP **37**(2–4). International Commission on Radiological Protection (2007).
14. Hille, R., Hill, P., Heinemann, K., Ramzaev, V., Barkovski, A., Konoplia, V. and Neth, R. *Current development of the human and environmental contamination in the Bryansk-Gomel Spot after the Chernobyl accident*. Radiat. Environ. Biophys. **39**, 99–109 (2000).
15. Zhu, Y. G. and Shaw, G. *Soil contamination with radionuclides and potential remediation*. Chemosphere **41**, 121–128 (2000).
16. Wang, X., Li, F., Okazaki, M. and Sugisaki, M. *Phytoremediation of contaminated soil*. Annual report from the Center for Environmental Science in Saitama, Vol. 3, pp. 114–123. Center for Environmental Science in Saitama (2003).

APPENDIX

Kerma-rates in air at 1 m above ground per unit activity per unit area (extracts from ICRU 1994) $(\mu Gy\ h^{-1})/(kBq\ m^{-2})$.

Radionuclide	Relaxation mass per unit area (β, g cm^{-2})[a]	
	1.0	2.0
^{131}I	1.12×10^{-3}	9.32×10^{-4}
^{134}Cs	4.44×10^{-3}	3.72×10^{-3}
^{137}Cs (^{137m}Ba)	1.73×10^{-3}	1.44×10^{-3}
^{129m}Te	1.28×10^{-4}	9.62×10^{-5}
^{95}Nb	2.17×10^{-3}	1.82×10^{-3}
^{136}Cs	5.90×10^{-3}	4.94×10^{-3}

[a]Set to 1.0 due to the passage of 3–6 weeks immediately after the accident at FNPP and 2.0 due to the passage of less than 1 year after the accident at FNPP.