ENVIRONMENTAL CONTAMINATION AND EXTERNAL RADIATION DOSE RATES

FROM RADIONUCLIDES RELEASED FROM THE FUKUSHIMA NUCLEAR POWER

PLANT

Yasuyuki Taira^{1,6}, Naomi Hayashida¹, Shunichi Yamashita², Takashi Kudo³, Naoki Matsuda⁴, Jumpei

Takahashi⁵, Alexander Gutevitc⁷, Alexander Kazlovsky⁸ and Noboru Takamura^{1*}

Departments of ¹Global Health, Medical and Welfare, ²Radiation Medical Science and ³Radioisotope

Medicine, Nagasaki University Graduate School of Biomedical Sciences, 1-12-4 Sakamoto, Nagasaki

852-8523, Japan

⁴Division 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

⁵Center for International Collaborative Research, Nagasaki University, 1-12-4 Sakamoto, Nagasaki

852-8523, Japan

⁶Nagasaki Prefectural Institute for Environmental Research and Public Health, 2-1306-11 Ikeda

Omura, Nagasaki 856-0026, Japan

⁷Zhitomir Inter-Area Medical Diagnostic Center, Kievskaya 21b, Korosten 260100, Ukraine

⁸Gomel State Medical University, 5 Lange Street, Gomel 240600, the Republic of Belarus

SHORT TITLE: EXTERNAL RADIATION DOSE RATES IN FUKUSHIMA

*Corresponding author: Noboru Takamura, M.D., Ph.D.

Professor and Chairman, Department of Global Health, Medical and Welfare

Nagasaki University Graduate School of Biomedical Sciences

1-12-4 Sakamoto, Nagasaki 852-8523, Japan

Phone: +81-95-819-7170: fax: +81-95-819-7172

E-mail: takamura@nagasaki-u.ac.jp

1

Abstract

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 analyzed by gamma spectrometry. Six artificial radionuclides (¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ^{129m}Te, ⁹⁵Nb, and ¹³⁶Cs) 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 (8.7-17.8 mSv/y) in Fukushima city on March 22, 2011. After several months, these calculated external effective doses were 0.25-0.88 μSv/h (2.2-7.6 mSv/y) in Fukushima city on June 29, 2011. The present study revealed that detected artificial radionuclides around FNPP mainly shifted to long-lived radionuclides such as radioactive cesium (¹³⁴Cs and ¹³⁷Cs) even though current levels are decreasing gradually due to the decay of short-lived radionuclides such as ¹³¹I, ^{129m}Te, ⁹⁵Nb, and ¹³⁶Cs. 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 March 11, 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 data of ¹³¹I and ¹³⁷Cs 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 approximately 10% (1.6×10¹⁷ Bq for ¹³¹I and 1.5×10¹⁶ Bq for ¹³⁷Cs) 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. Accessed February 25, 2012).

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 ingestion of contaminated foods produced in contaminated areas. It is extremely important to evaluate the environmental contamination and 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 foods intake by the nation are strictly carrying out after the FNPP accident (http://www.mhlw.go.jp/english/topics/2011eq/index.html. accessed February 25, 2012). 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 analyzed by gamma spectrometry. Furthermore,

external	effective	doses	were	calculated	from	samples	from	these	areas	for th	e estimation	ı of	radiation
exposur	e status.												

Materials and Methods

Sampling places

Fukushima Dai-ichi Nuclear Power Plant is located on the east coast of the island of Honshu, approximately 200 km northeast of Tokyo. Samples around FNPP were collected from: Fukushima city (the prefectural capital, N37° 41′, E140° 28′), 57.8 km northwest of FNPP (N37° 25′, E141° 02′); Iwaki city (N37° 03′, E140° 53′), 43.4 km south of FNPP; Ono town (N37° 17′, E140° 37′), 39.1 km southwest of FNPP and litate village (N37° 40′, E140° 44′), 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 cm and 5-10 cm) around FNPP between March and July, 2011 (Fukushima city on March 22 and June 29, Iwaki city on April 7, Ono town on April 7 and Iitate village on July 11, 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 analyzed with a high purity germanium detector (ORTEC®, GEM35, Ortec International Inc., Oak Ridge, TN, USA) coupled to a multi-channel analyzer (MCA7600, Seiko EG&G Co., Ltd., Chiba, Japan) for 7,200 s (FNPP). We set the measuring time for that at which objective radionuclides could be detected sufficiently, although we must reinforce monitoring of the environmental after the FNPP accident. Gamma-ray peaks used for measurements were 364.48 keV for ¹³¹I, 604.66 keV for ¹³⁴Cs, 661.64 keV for ¹³⁷Cs, 695.98 keV for ^{129m}Te, 765.79 keV for ⁹⁵Nb and 818.50 keV for ¹³⁶Cs. 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 authorized by the Ministry of Education, Culture, Sports, Science, and Technology, Japan (http://www.kankyo-hoshano.go.jp/en/index.html. Accessed February 25, 2012) (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:

$$H_{ext} = C \cdot D_{ext} \cdot f \cdot s \tag{1}$$

in which C is the activity concentration of detected artificial radionuclides (kBq/m²; calculated from radionuclide concentration in Bq/kg 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 [(μ Gy/h)/(kBq/m²)] for detected artificial radionuclides with the relaxation mass per unit area (β : g/cm²) 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 less than 1 year after the accident at FNPP (dose conversion coefficients are shown in the Appendix) (²); f is the unit conversion coefficient (0.7 Sv/Gy for effective dose rate in the body per unit absorbed dose rate in air) (³); and 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 cesium (¹³4Cs and ¹³7Cs) were calculated using Eq. (1) (H_{ext} ×24h×365d).

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 ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ^{129m}Te, ⁹⁵Nb, and ¹³⁶Cs (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 ¹³⁴Cs, ¹³⁷Cs, 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 these 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/m² of detected artificial radionuclides in surface soils (0-5 cm) around FNPP were calculated from these radionuclides concentrations in Bq/kg (these concentrations are shown in Table 3).

The external effective doses from detected artificial radionuclides around FNPP using Eq. (1) are summarized in Table 4. Immediately after the accident, calculated external effective doses around FNPP were 1.9-2.9 μ Sv/h (8.7-17.8 mSv/y) in Fukushima city, 0.058 μ Sv/h (0.092 mSv/y) in Iwaki city, and 0.092 μ Sv/h (0.62 mSv/y) in Ono town, respectively. Several months after the accident, the calculated external effective doses around FNPP were 0.25-0.88 μ Sv/h (2.2-7.6 mSv/y) in Fukushima city and 0.53 μ Sv/h (4.6 mSv/y) 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 μ Sv/h around 30 km over the distance from the reactor (Fukushima city, Iwaki city, and Ono town) from March 29 to April 8, 2011 and 0.17 μ Sv/h 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 March 12, 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 localized areas, these accumulated volumes of radioactive materials are at high levels. Thus, these emergency zones are extremely important areas for practicing 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. Accessed February 25, 2012) such as litate 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 ¹³¹I, ^{129m}Te, ⁹⁵Nb, and ¹³⁶Cs, 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 (Table 1 and Table 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 radionuclides 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 ¹³⁷Cs deposition density greater than 555 kBq/m² (15 Ci/km²) are designated as areas of strict control about the CNPP accident on April 26, 1986⁽³⁾. Long-term (10 days) 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-cesium, 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 ¹³¹I ⁽³⁾. Deposition in the nearby contaminated zone (<100 km) reflected the radionuclide composition of the fuel and the volatile elements including iodine and cesium in the form of condensation-generated particles were more

widely dispersed into the far zone (from 100 km to approximately 2,000 km) (3). The ¹³⁷Cs deposition was highest within a 30-km radius area surrounding the reactor, known as the 30-km zone, and deposition densities exceeded 1,500 kBg/m² 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/m² (0.11 Ci/km²) from April to July 1986 (6). In 2000, the total inventories of the fuel component 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 about 1.2 times higher, and internal doses were about 1.1-1.5 times higher, than those obtained during 1986-1995 (depending on soil properties and applied countermeasures) (7). Moreover, our previous study revealed that areas around CNPP, especially the Russian-Belarussian border including Klincy city and Gomel oblast, are still contaminated (8). Although releasing amounts of artificial radionuclides from nuclear reactors and diffusion scales differ between FNPP and CNPP, data of the environmental radioactivity level around CNPP is 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 ¹³¹I and ¹³⁴Cs. At several months after the accident, detected artificial radionuclides around FNPP mainly shifted to radioactive cesium (¹³⁴Cs and ¹³⁷Cs, Table 1). In general, the volatile elements primarily consist of radioactive iodine, including ¹³¹I, which has a radioactive half-life of 8 days, and radioactive cesium, including ¹³⁷Cs with a half-life of 30 years. 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 ¹³¹I are decaying gradually (http://www.mext.go.jp/english/. Accessed February 25, 2012). In the CNPP accident, more than 4,000 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, Ukraine, and the four most contaminated regions of Russia ⁽⁹⁾. Most of the thyroid dose was caused by the intake of ¹³¹I 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 cesium of Fukushima city and litate village located in northwest of FNPP exceeded 1mSv/y, the public dose limit (ICRP, 1991) (Table 4). The national government aims to reduce the estimated annual exposure dose to less than 20 mSv on the areas with an estimated annual exposure dose of greater than 20 mSv (emergency exposure situations) and the estimated annual exposure dose closer to 1 mSv on the areas with an estimated annual exposure dose of less than 20 mSv with municipalities and local residents to conduct effective decontamination work (existing exposure situations) (12). 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 fundamental data for health effects by chronic and low level exposure such as radioactive cesium, especially ¹³⁷Cs.

There were several limitations in this study. Several radionuclides could not be analyzed by an extraction procedure, including ⁹⁰Sr, and homogenized soil samples in extensive areas could not be standardized. 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 ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs were detected at high levels in the areas around FNPP immediately after the accident, these radionuclides could be analyzed 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 (13). However, it was reported that reduced plant uptake of radionuclides, especially 137Cs and 90Sr, by competitive cations contained in chemical fertilizers has a general advantage in large-scale, low-level contamination incidents on arable land and has been widely practiced in central and western Europe after the Chernobyl accident (14). The existing remediation approaches and phytoextraction (phytoremediation) of radionuclides from contaminated soils have been examined (15). 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 to other areas around FNPP and contributed to the comparatively short-lived radionuclides such as ¹³¹I and ¹³⁴Cs. At several months after the accident, detected artificial radionuclides around FNPP mainly shifted to radioactive cesium (¹³⁴Cs and ¹³⁷Cs). 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, 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.

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.

Acknowledgements

We would also like to thank Mr. Shinichi Nakamura and Mr. Kohji Yoshida for assistance with environmental sampling and monitoring around the Fukushima Nuclear Power Plant.

References

- Ministry of Education, Culture, Sports, Science, and Technology, Japan. *Environmental Radioactivity and Radiation in Japan* (http://www.kankyo-hoshano.go.jp/en/index.html. Accessed February 25, 2012)
- International Commission on Radiation Units and Measurements. Gamma-Ray Spectrometry in the Environment. ICRU Report 53 (Bethesda, Maryland: International Commission on Radiation Units and Measurements) (1994)
- United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. 2000 Report to General Assembly, with scientific annexes. (New York: United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations) (2000)
- 4. Kulan, A. Seasonal ⁷Be and ¹³⁷Cs activities in surface air before and after the Chernobyl event. J. Environ. Radioact. 90, 140-150 (2006)
- 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)
- 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 ¹³⁷Cs 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: Bennet B, Repacholi M and Zhanat C eds. Report of the UN Chernobyl Forum expert group "Health". pp. 23-53. (Geneva: World Health Organization) (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., Khrouch, V., Shinkarev, S., Drozdovitch, V., Minenko, V., Shemiakina, E., Ulanovsky, A., Bouville, A., Anspaugh, L., Voillequé, P. and Luckyanov, N. *Individual thyroid dose estimation for a case-control study of Chernobyl-related thyroid cancer among children of Belarus. Part 1:*131 I, short-lived radioiodines (132 I, 133 I, 135 I), and short-lived radiotelluriums (131 m Te and 132 Te).

 Health Phys. 86, 565-585 (2004)
- 12. International Commission on Radiological Protection. *The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103; Annals of the ICRP, vol. 37, Nos. 2-4* (Stockholm: International Commission on Radiological Protection) (2007)
- 13. 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)
- 14. Zhu, Y.G. and Shaw, G. Soil contamination with radionuclides and potential remediation.

 Chemosphere 41, 121-128 (2000)
- 15. Wang, X., Li, F., Okazaki, M. and Sugisaki, M. *Phytoremediation of contaminated soil*. Report from the Center for Environmental Science in Saitama 3, 114-123 (2003)

Figure 1. Areas around the Fukushima Nuclear Power Plant

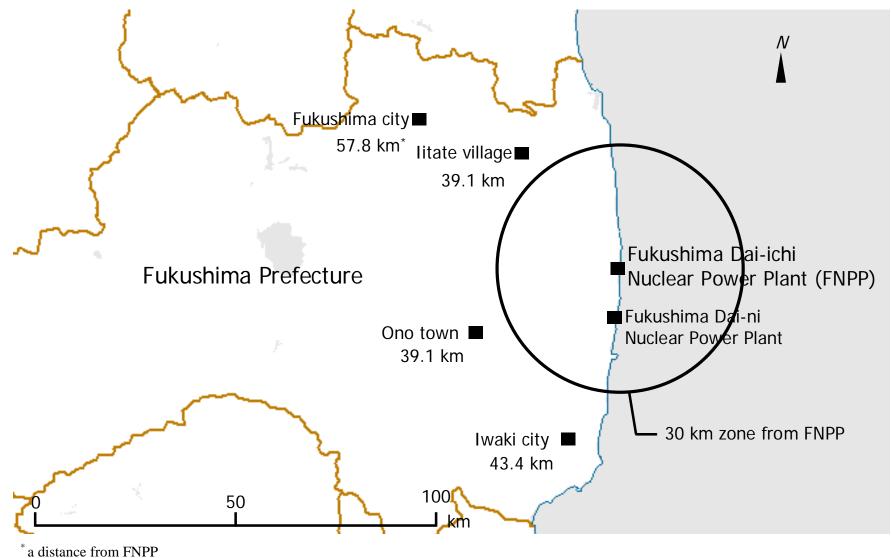


Table 1. Distribution (Bq/kg-dry) of detected artificial radionuclides in surface soils (0-5 cm) collected at areas around Fukushima Nuclear Power Plant

	Radionuclide concentration in Bq/kg-dry						
_	¹³¹ I	¹³⁴ Cs	¹³⁷ Cs	^{129m} Te	⁹⁵ Nb	¹³⁶ Cs	
	$(8.0d)^{a}$	(2.1y)	(30y)	(34d)	(35d)	(13d)	
Fukushima city A (March)	54150±780 ^b	13402±85.6	12989±81.9	14908±1017	n.d. ^d	1649±106	
rukusiiiiia city A (waicii)	$(1410)^{c}$	(71)	(51)	(2390)	(78)	(241)	
Fukushima city B (March)	34287 ± 670	20838±93.1	20427 ± 89.5	20288±1137	n.d.	2521±116	
Tunusiiiiia erey 2 (iviai eii)	(1545)	(77)	(57)	(2797)	(82)	(258)	
Fukushima city C (March)	18775 ± 429	11417±59.8	11158±57.1	10654 ± 721	75.5 ± 18	1319±74	
Tukusiiiiia eity C (Maieii)	(991)	(51)	(36)	(1800)	(51)	(171)	
Fukushima city D (June)	n.d.	26560 ± 70.2	28690±73.5	2531±449	n.d.	n.d.	
rukusiiiiia eity D (Julie)	(313)	(57)	(43)	(1312)	(40)	(72)	
Fukushima city E (June)	n.d.	6107 ± 27.5	6648 ± 29.0	n.d.	n.d.	n.d.	
rukusiiiiia eity E (Julie)	(129)	(24)	(18)	(582)	(18)	(30)	
Fukushima city F (June)	n.d.	12836±32.5	14257±34.9	1712±254	n.d.	n.d.	
Tukusiiiiia city i (Julie)	(162)	(27)	(21)	(738)	(22)	(41)	
Iwaki city (April)	3560 ± 33.2	146 ± 4.2	152 ± 4.4	631±73	n.d.	11.4 ± 2.7	
Iwaki city (Apiii)	(23)	(6.2)	(5.4)	(169)	(6.6)	(7.7)	
Ono town (April)	1403 ± 26.2	1052±10.6	1099±11.1	672±98	n.d.	60.2 ± 5.7	
Ono town (April)	(44)	(9.6)	(7.8)	(264)	(7.7)	(13)	
Iitate village (July)	n.d.	12799 ± 41.8	14151±45.3	1629 ± 251	n.d.	n.d.	
Thate viriage (July)	(77)	(34)	(28)	(725)	(21)	(25)	

^a half-life

Samples were collected at areas around FNPP between March and July, 2011 (Fukushima city on March 22 and June 29, Iwaki city on April 7, Ono town on April 7 and Iitate village on July 11). Radionuclides were analyzed with a germanium-detector coupled to a multi-channel analyzer for 7,200 s at Nagasaki Prefectural Institute for Environmental Research and Public Health, Nagasaki, Japan.

^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 cm and 5-10 cm) collected at areas around Fukushima Nuclear Power Plant

Radionuclide concentration in Bq/kg-dry 131 **I** ^{129m}Te 95Nb ¹³⁶Cs ¹³⁴Cs 137 Cs $(8.0d)^{a}$ (2.1y)(30y)(34d)(35d)(13d)3560±33.2^b 146±4.2 152±4.4 631±73 n.d. d 11.4 ± 2.7 Iwaki city (0-5 cm) (April) $(23)^{c}$ (6.2)(5.4)(169)(6.6)(7.7)2219±30.8 138 ± 4.3 148 ± 4.5 310±69 n.d. n.d. Iwaki city (5-10 cm) (April) (26)(5.2)(4.6)(194)(7.0)(16) 1403 ± 26.2 1052±10.6 1099±11.1 672 ± 98 n.d. 60.2 ± 5.7 Ono town (0-5 cm) (April) (44) (9.6)(7.8)(264)(7.7)(13) 325 ± 14 217±5.1 223±5.4 n.d. n.d. n.d. Ono town (5-10 cm) (April) (5.0)(5.3)(172)(26)(5.1)(16)n.d. n.d. 12799±41.8 14151±45.3 1629 ± 251 n.d. Iitate village (0-5 cm) (July) (77)(34)(28)(725)(21) (25)5788±30.7 5256±28.7 n.d. n.d. n.d. n.d. Iitate village (5-10 cm) (July) (58)(24) (20)(481)(15)(19)

Samples were collected at areas around FNPP between April and July, 2011 (Iwaki city on April 7, Ono town on April 7 and Iitate village on July 11). Radionuclides were analyzed with a germanium-detector coupled to a multi-channel analyzer for 7,200 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 3. Distribution (kBq/m²) of detected artificial radionuclides in surface soils (0-5 cm) collected at areas around Fukushima Nuclear Power Plant

	Radionuclide concentration in kBq/m ²						
_	¹³¹ I (8.0d) ^a	¹³⁴ Cs (2.1y)	¹³⁷ Cs (30y)	^{129m} Te (34d)	⁹⁵ Nb (35d)	¹³⁶ Cs (13d)	
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 Fukushima Nuclear Power Plant

	External effective dose		Mainly contributed radionuclides		
	μSv/h *	mSv/y [†]			
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		
litate village (July)	0.53	4.6	¹³⁴ Cs, ¹³⁷ Cs		
over 30 km distance from FNPP reactor (March-to-April)	0.39-0.41 ‡				
over 30 km distance from FNPP reactor (July)	0.17				

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/m²; calculated from radionuclide concentration in Bq/kg 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 ((μ Gy/h)/(kBq/m²) for detected artificial radionuclides with the value of relaxation mass per unit area 1.0 and/or 2.0 g/cm² (ICRU 1994)), f is the unit conversion coefficient (0.7 Sv/Gy (UNSCEAR 2000)), f is the occupancy-shielding factor (0.2 fractional time outdoors + 0.8 fractional time indoors × 0.2 building shielding = 0.36 (UNSCEAR 2000)).

[†]calculated external effective doses due to radioactive cesium (134Cs and 137Cs)

[‡]external effective doses were estimated from a portable detector of monitoring personnel who sampled soils around FNPP (PDM-112[®], Hitachi-Aloka Medical, Ltd., Tokyo, Japan)

Appendix. Kerma-rates in air at 1 m above ground per unit activity per unit area (extracts from ICRU 1994)

		$(\mu Gy/h)/(kBq/m^2)$			
Radionuclide ——	Relaxation mass per unit area (β: g/cm²) *				
Radionucide	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}			

^{*} 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