Measurement of the environmental radiation dose due to the accident at the Fukushima Daiichi Nuclear Power Plant

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On March 11, 2011, an undersea megathrust earthquake caused a tsunami that inflicted serious damage to the Fukushima Daiichi Nuclear Power Plant (FDNPP). On March 12, 2011, we began measuring environmental radiation doses and identifying fission product radionuclides at the International University of Health and Welfare (IUHW). The purpose of this investigation is to estimate the external exposure dose of fission products from FDNPP. Measurements were performed between March 12 and August 31, 2011. A NaI(Tl) scintillation survey meter was used to measure the environmental radiation dose, and air dust samplers and a NaI(Tl) scintillation spectrum analyzer were used to identify radionuclides in the atmosphere and soil. For estimating external doses, three lifestyle groups were considered viz. students or office workers, business persons, and farmers or construction workers. Increasing doses were detected on March 15 around noon, and the doses peaked on March 16. Post-peak, the doses decreased exponentially and became stable after two months. Immediately after the accident, some fission product radionuclides were detected in the atmosphere and in the soil samples. Almost no radionuclides were detected in the atmosphere approximately one month after the first analysis; although the radiation was decaying, radionuclides were detected in the soil and were isolated. The external dose varied with the supposed lifestyle; assuming that the abundance ratio of Cs-134 to Cs-137 was 1:1, the annual external doses for the considered lifestyles were 1.069 mSv for students or office workers, 1.672 mSv for business persons, and 2.044 mSv for farmers or construction workers. These doses are sufficiently small so that most residents including children, living near IUHW, would not be affected. Further investigation of the internal exposure is necessary for a better estimate of the effective doses. External exposure to fission product radionuclides is within safe levels, and while further investigation of internal exposure factors such as milk, water, and mushrooms is still needed, it appears that radiation around IUHW does not pose a health hazard.

Key Words : *radiation survey, radiation dose, external dose, Fukushima, fission product*

1. Introduction

At 14:46 on March 11, 2011, an earthquake of unprecedented magnitude centered off the Pacific coast of Japan's Tohoku region induced a massive tsunami that surged over the coast, including the Fukushima Daiichi Nuclear Power Plant (FDNPP)[1]. Most power systems at FDNPP were destroyed, and as a result, four nuclear reactors and many tanks for storing nuclear fuels could not be controlled. The nuclear reactors and fuel could not be cooled, thereby inducing meltdowns that increased pressure in the nuclear reactor and the water temperature in the cooling tanks. The situation became increasingly serious, to the point where the outer walls of reactors 1 and 3 were blown off by hydrogen explosions. Anticipating a possible explosion of the nuclear reactors would explode, at around noon on March 12, FDNPP workers decided to open vents to relieve the high pressure in the reactors. This released massive amounts of fission products into the atmosphere, increasing the environmental radiation dose throughout the Kanto and Tohoku regions. Having predicted that many fission products would be dispersed in the immediate aftermath of the tsunami, we began measuring environmental radiation doses around the International University of Health and Welfare (IUHW) on March 12 and 13. Commencing from March 14, we also performed fixed-point measurements of the environmental radiation doses at three locations viz. indoors, on asphalt, and on the ground.

Classified as level 7 on the international nuclear event scale, the Fukushima nuclear accident is one of the worst in history. In comparison to the similarly rated Chernobyl disaster, the amounts of radionuclides released from FDNPP was lower due to differences in the types of nuclear fuels used and in the various counter measures taken after the accident. Nonetheless, it is likely that radiation exposure due to the enormous amount of fission products released cannot be avoided in the long term. There have been epidemiological surveys of the effects of radiation exposure following the accident at Chernobyl in Ukraine (April 26, 1986) and of atomic bomb survivors with radiation sickness in Hiroshima and Nagasaki (August 6 and 9, 1945)[2-10]. These studies facilitate some predictions of the effects of radiation exposure on human health.

The purpose of this investigation is to evaluate the fission product radionuclides and the expected external radiation dose in lifestyle groups, from environmental radiation doses measured around Otawara City in Tochigi Prefecture, approximately 110 km away from FDNPP, starting the day following the accident.

2. Method

(1) Identification of radionuclides

Identification of fission product radionuclides in the atmosphere and in soil samples was performed twice on different dates. Dust suspended in the atmosphere was collected on March 16 and April 8, 2011, and soil was collected on March 16 and April 12, 2011. Dust was collected for a period of 60 min, by using a dust sampler (DSM-205, Hitachi Aloka Medical Ltd., Tokyo) and filter paper (60 mm, HE40T, Advantec, Tokyo), outdoors in the vicinity of IUHW, 110 km from the FDNPP (Fig. 1). Three 100 g soil samples were collected from two areas with shrubbery near IUHW, and were mixed well. For testing, 40 g of the mixture was used. To identify radionuclides, samples were analyzed using a NaI(Tl) scintillation detector (6S6P1.5C2, Radiation Sensors LLC, USA) and a spectral analyzer (UCS-30, Spectrum Techniques, USA), which were calibrated using a sealed cesium-137 source as a standard source. Lead blocks (5 cm × 5 cm × 10 cm) were used to shield the detector on the NaI(Tl) scintillation spectral analyzer from background radiation. The measurement time was 10 min for all samples.

(2) Measurement of environmental radiation dose

We began measuring the environmental radiation dose with a NaI(Tl) scintillation survey meter (TDS-161, Hitachi Aloka Medical Ltd., Tokyo, Japan) on March 12, 2011, at three locations around IUHW viz. indoors, on

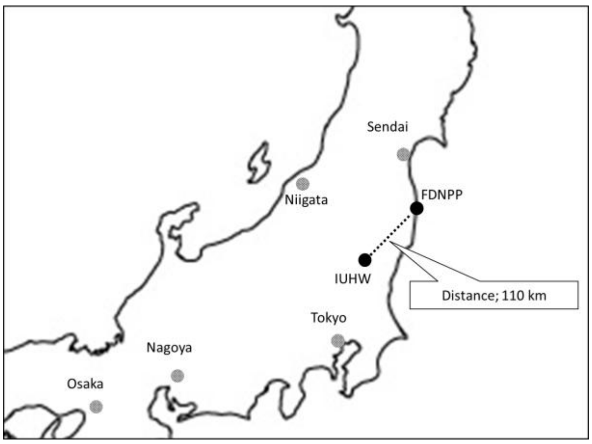


Fig. 1. Location of IUHW in Otawara City and FDNPP

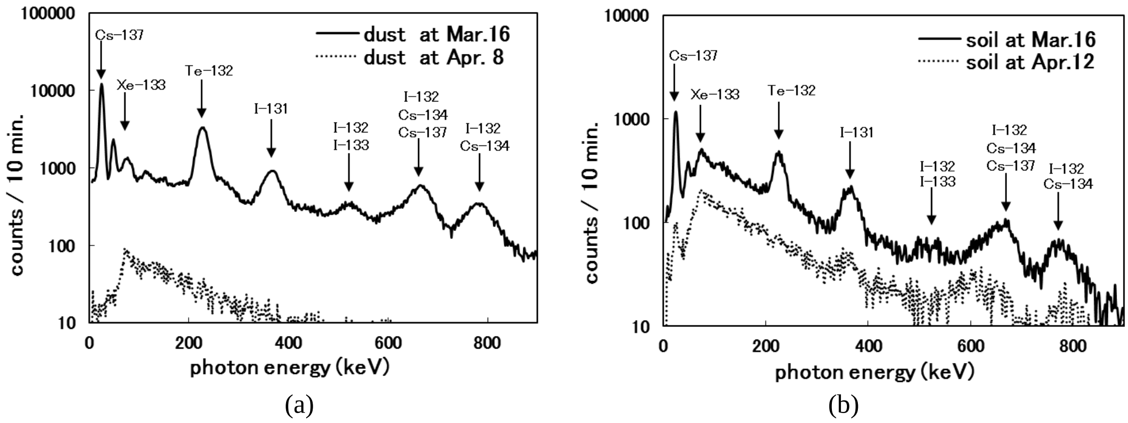


Fig. 2. Energy spectrum of radionuclides in the atmosphere and in soil.
(a) In the atmosphere: the continuous line is the spectrum on March 16, and the dotted line is the spectrum on April 8.
(b) In soil: the continuous line is the spectrum on March 16, and the dotted line is the spectrum on April 12.

asphalt, and on the ground. On March 12 and 13, we measured radiation doses 6 times per day at approximately 1–2 h measurement intervals between 9:00 hrs and 17:00 hrs. Between March 14 and April 30, measurements were performed hourly between 9:00 hrs and 16:00 hrs, and subsequently once per day until October 31. Presently, environmental radiation doses are measured once per week. The height of the center of the detector from the ground was fixed at 1 m, and steady digital values were recorded after preliminary operation for 2 min. The time constant was 10 s in all cases. To fill gaps in the data for which measurements were not performed, such as during stormy weather, the linearly interpolated value of the previous day and the subsequent day has been used.

(3) Estimation of external radiation dose

Estimations of external radiation doses were calculated for three lifestyle cases (Table 1). The time spent at home (60% doses of that on asphalt) was assumed to be 12 h for each case. Case 1 assumed an office worker or student, spending 3 h on asphalt and 9 h indoors (40% doses of that on asphalt). Case 2 assumed a worker outside the office, spending 9 h on asphalt and 3 h on the ground. Case 3 was assumed a farmer or construction worker, spending 3 h on asphalt and 9 h on the ground. In these estimations, the maximum value for each day was used in the calculations from March 14 to April 30, and was assumed to be constant during the day. Between May 1 and August 30, the measured value at 12:00 noon was assumed to be the constant value for the day. After September 1, the accumulated dose of external radiation

Table 1. Three case lifestyles, assuming various times spent at home, indoors, on asphalt, and on the ground.

	home (60% of on asphalt)	indoor (40% of on asphalt)	on asphalt	on ground
Case - 1	12 h	0 h	3 h	9 h
Case - 2	12 h	0 h	9 h	3 h
Case - 3	12 h	9 h	3 h	0 h

for the year since March 14 was calculated by using the estimated virtual external radiation doses according to the decay of cesium-134 and cesium-137.

3. Result

(1) Identification of radionuclides

On March 16, iodine-131, iodine-132, iodine-133, tellurium-132, xenon-133, cesium-134, and cesium-137 were identified in the energy spectrum of radionuclides in the atmosphere; but radionuclides were not identified on April 8 because the measured values were below 100 counts (Fig. 2a). The same radionuclides as those found in the atmosphere were also detected in the soil on March 16. On April 12, however, the measured counts were below those on March 16, and iodine-132, tellurium-132, and iodine-133, having half-lives of 20.8 h, 3.2 days, and 2.3 h, respectively were not detected (Fig. 2b)

(2) Changes in environmental radiation dose

Figure 3 shows changes in the environmental radiation dose around IUHW between March 12 and April 30,

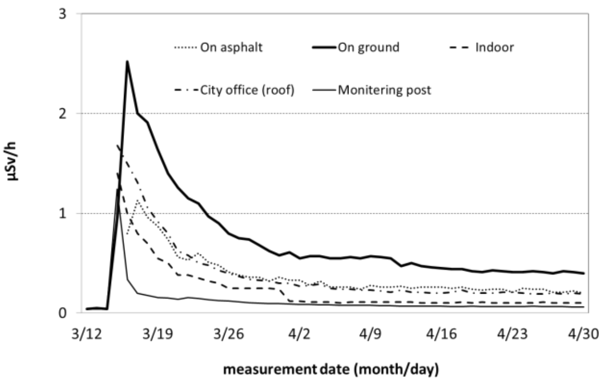


Fig. 3. Changes in environmental radiation dose around IUHW between March 12 and April 30.

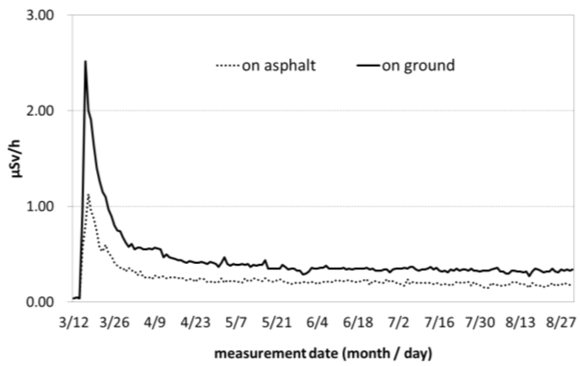


Fig. 4. Changes in environmental radiation doses at two locations in IUHW between March 14 and August 30.

together with measurements at the nearest monitoring post and city office. The background radiation around IUHW before the accident was approximately 0.04 to 0.06 μSv/h. A sudden increase in radiation was detected at noon on March 15, with a maximum value of 2.52 μSv/h on the ground on March 16, due to rain on the night of the first detection. Subsequently, the radiation dose decreased exponentially to an approximately constant value of 0.35 μSv/h on the ground two months later. Official measurement data from the Utsunomiya monitoring post shows a decrease immediately after the first detection, but the measurement data from the city office in nearby Nasu Town is similar to our data on asphalt. On April 1, a detector near a window was moved to the center of the room, resulting in lower readings. Figure 4 shows differences in the environmental radiation dose in asphalt and on the ground at IUHW between March 12 and August 30. A moderate decrease in radiation dose was observed on and after May 1, because various radionuclides with short half-lives varied from this trend. Therefore, the trend is determined by radionuclides with medium half-lives, such as cesium-134 (2.2 y). Following this, cesium-137 (30 y) would most heavily influence the decrease.

The continuous thick line shows values on the ground, the dotted line shows values on asphalt; the dashed line shows values indoors; the continuous thin line shows values from the monitoring post, and the dashed/single-dotted line is the data from the city office nearest IUHW.

(3) Estimation of external radiation dose

For the environmental radiation doses, the annual external radiation doses since March 14 were estimated under the assumption that radiation doses will decrease according to the half-lives of cesium-134 and cesium-137 (whose abundance ratio was assumed to be 1:1). These doses were calculated for cases 1 to 3, and estimated as the external radiation doses over one year (Fig. 5). Case 1, case 2, and case 3 were predicted to receive 1.069, 1.672, and 2.044 mSv/y, respectively. The estimated external radiation doses were influenced by the time spent on unpaved ground.

4. Discussion

Ideally, a semiconductor detector system is best suited to analyze radionuclides, but we used a NaI(Tl) scintillation detector system because our germanium detector system was not operational at that time. A NaI(Tl) scintillator detector is not sufficiently precise for analysis, but this system was sufficient in identifying these radionuclides as fission products soon after the accident. Cesium-134, cesium-137, xenon-133, tellurium-132, and others were thus detected and identified using this system, proving that these radionuclides were fission products from FDNPP. The identification of radionuclides in atmospheric dust was difficult under the same conditions approximately one month later. This was expected because radionuclides in the atmosphere fell to the ground with rain on the night of March 15 as evident from the various radionuclide decay products which could be identified in soil but not in the atmosphere, while some short half-life nuclides such as iodine-131 or tellurium-132 were not detected in the soil samples. The NaI(Tl) scintillation detector was sufficiently useful for this investigation in the early phase because, in short, many radionuclides in the atmosphere were absorbed into the soil, and nuclides with short half-life were not detected from their decay[11].

We first used the NaI(Tl) scintillation survey meter, an

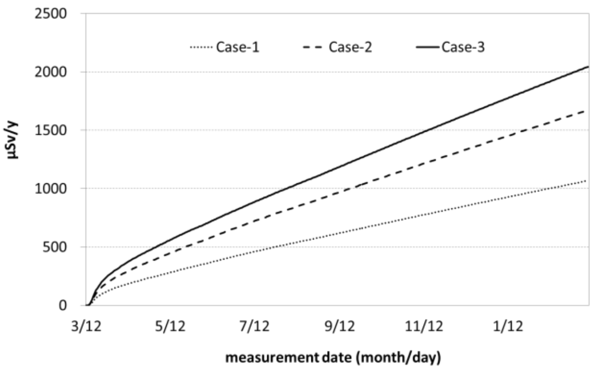


Fig. 5. Estimation of external radiation doses over one year for cases 1, 2, and 3.
The dotted line shows case 1, the dashed line shows case 2, and the continuous line shows case 3.

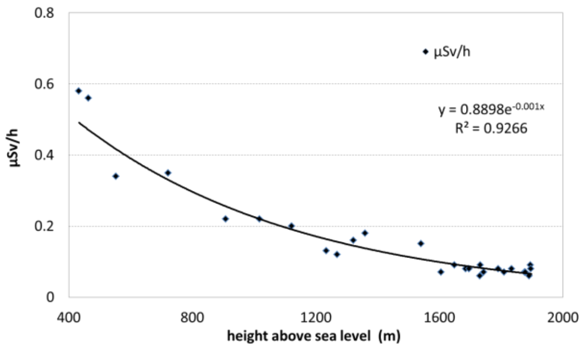
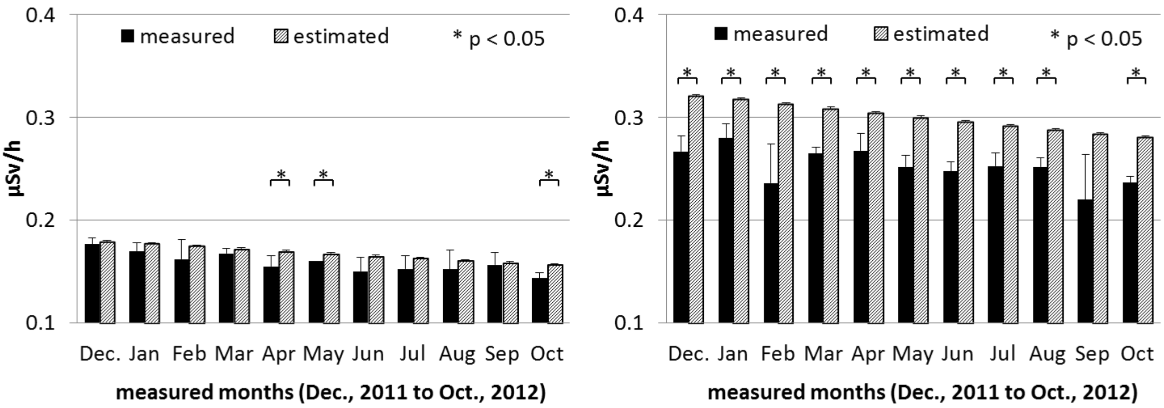


Fig. 6. Environmental radiation dose at Mt. Nasu, 1,915 m above sea level.



(a) (b)

Fig. 7. Comparison of measured and estimated external radiation doses between August 1, 2011 and October 24, 2012:
(a) on asphalt- solid bar is measured data, shaded bar is estimated data
(b) on ground- solid bar is measured data, shaded bar is estimated data

ionization chamber survey meter, and a Geiger-Mueller survey meter to measure the environmental radiation doses, but the NaI(Tl) scintillation survey meter was adopted due to concerns about changes in radiation doses. The maximum value was recorded on March 16 due to rain on the night of March 15, but doses decreased exponentially after that and stabilized after two months. Asphalt and ground measurement values on March 16 were 0.80 and 2.52 µSv/h, respectively, and on April 1 had decreased to 0.33 and 0.61 µSv/h, respectively. The reason for this could be that the decay of iodine-131 (half-life: 8 days) was the dominant factor. Therefore, the radionuclides contributing to the radiation dose could be estimated as cesium-134 or cesium-137 i.e. nuclides with medium or long half-lives exceeding 2 years.

Mt. Nasu extends 1,915 m above sea level and is covered by sediment from volcanic eruptions. The mountain has little vegetation over an altitude of 1,700 m. We performed measuring the environmental radiation dose at Mt. Nasu using by the same survey meter and method for measurements in this study. The radiation dose decreased in inverse proportion to the altitude ($y=0.8898e^{-0.001x}$, $R^2=0.9266$), and the dose at the top there was approximately 0.06 µSv/h on August 20, 2011 which was almost the same as the background radiation before the accident, but accumulations of radionuclides with approximately 0.20 to 0.59 µSv/h were detected in vegetated areas (Fig. 6). This result suggests that environmental radiation doses depend more on the amount of soil than on altitude above sea level.

Our measurement of radiation doses was completed on August 31, and then restarted at intervals of once per week beginning on December 14, 2011. The estimated radiation doses were compared with measured values from August 1, 2011, to October 24, 2012 (Fig. 7). The doses on asphalt were almost unchanged, but the doses on the ground had decreased slightly (paired t-test, $p<0.05$). Accordingly, there were a few differences between estimated and measured values.

We predict that time spent on unpaved terrain such as ground or farms influences the external radiation dose for each lifestyle, and that the dose will be less than 2.1 mSv/y. Such levels of radiation are not injurious to the health of residents living near IUHW, but internal radiation doses were not considered in this investigation. Takatsuji et al. reported that internal radiation dose depends on diet[5]. Internal radiation doses are assumed to be lower than those that were seen after Chernobyl, because strict tests for food radiation have been performed, and because wild mushrooms are not widely consumed; however, investigation should continue for foods that are a source of concern in relation to radionuclide contamination. Internal radiation exposure by respiration is another possible factor, and we should pay special attention to the internal radiation dose to infants from food and respiration

5. Conclusion

We have measured environmental radiation doses since the day following the nuclear accident at FDNPP, and found that many fission product radionuclides fell from the atmosphere to the ground and accumulated in the soil. We predict that there is no health hazard, because the external radiation exposure for multiple lifestyles was estimated to be 1.1–2.1 mSv/y. The effective radiation doses should nonetheless be evaluated, considering both external and internal radiation exposure.

References

1) Prime Minister of Japan and His Cabinet, Report of Japanese Government to the IAEA Ministerial Conference on Nuclear Safety -The Accident at TEPCO's Fukushima Nuclear Power Stations, 2011 (http://www.kantei.go.jp/foreign/kan/topics/201106/iaea_houkokusho_e.html)
2) Report of the Chernobyl Forum Expert Group ‘Environment’, Environmental Consequences of The Chernobyl Accident and Their Remediation; Twenty Years of Experience, International Atomic Energy Agency, 2006
3) I A Zvonova, T V Jeskot, M I Balonov, et al, 134Cs and 137Cs Whole-Body Measurements and Internal Radiation Dosimetry of The Population Living in Area Contaminated by Radioactivity After The Chernobyl Accident, Radiation Protection Dosimetry, 62(4), 213-221, 1995
4) N Hayashida, Y Sekitani, A Kozlovsky, et al, Screening for 137Cs Body Burden Due to the Chernobyl Accident in Korosten City, Zhitomir, Ukraine: 1996–2008, J. Radiat. Res., 52, 629-633, 2011
5) T Takatsuji, H Sato, J Takada, et al, Relationship Between The 137Cs Whole-Body Counting Results and Soil and Food Contamination in Farms Near Chernobyl, Health Phys., 78(1), 86-89, 2000
6) M Hoshi, Y O Konstantinov, T Y Evdeeva, et al, Radiocesium in Children Residing in The Western Districts of The Bryansk Oblast from 1991-1996, Health Phys., 79(2), 182-186, 2000
7) C Thornberg, R Vesanen, E Wallsrom, et al, External and Internal Irradiation of a Rural Bryansk (Russia) Population from 1990 to 2000, Following High Deposition of Radioactive Cesium from the Chernobyl Accident, Radiat Environ Biophys, 44, 97-106, 2005
8) M P Little, P Hall, M W Charles, Are Cancer Risks Associated with Exposures to Ionizing Radiation from Internal Emitters Greater than those in the Japanese A-bomb Survivors?, Radiat Environ Biophys, 46, 299-310, 2007
9) J C Kaiser, P Jacob, R Meckback, et al, Breast Cancer Risk in Atomic Bomb Survivors from Multi-Model Inference with Incidence Data 1958–1998, Radiat Environ Biophys, 23, Published online 1-14,2011
10) Y Shimizu, H Kato, W J Schull, Risk of Cancer among Atomic Bomb Survivors, J Radiat. Res., 2, 54-63, 1991
11) Report of the Chernobyl Forum Expert Group ‘Environment’, Environmental Consequences of the Chernobyl Accident and their Remediation: Twenty Years of Experience, IAEA, 2006