Comparison of fallout in rain due to the Fukushima and Chernobyl reactor accidents and the Hiroshima atomic bomb

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The fallout in rain due to the Fukushima reactor accident was monitored for about two months (from March 20 to May 23, 2011) in Higashi-Hiroshima city, Hiroshima, Japan. Gamma-ray spectra measured with a low background HPGe spectrometer showed clear evidence of the following fission products: ^{131}I, ^{137}Cs, and ^{134}Cs. ^{131}I was observed on March 27 and April 8, while ^{137}Cs and ^{134}Cs were observed on March 27, April 18, and April 22. The ^{131}I, ^{137}Cs, and ^{134}Cs activity concentrations in rainwater collected in Hiroshima reached 0.44 Bq/L on April 8, 0.17 Bq/L on April 18, and 0.15 Bq/L on April 18, respectively. Several samples of rainwater collected in Chiba (Kashiwa) on March 21, April 11, and May 12, Tokyo (Nerima) on March 21 and April 11, Osaka (Hirano) on April 8, Nara (Kitakatsuragi) on April 9, and Fukushima (Fukushima) on April 19, were also measured using our spectrometer. Our results from the Fukushima reactor accident were compared with measurements taken after the Chernobyl reactor accident as well as analyses of the black rain that was caused by fallout from the Hiroshima atomic bomb.

Key Words : *Radioactive fallout, Fukushima accident, Fission product, Gamma spectrometry*

1. Introduction

The spread/distribution of radioactivity caused by the accident at TEPCO's Fukushima Daiichi Nuclear Power Station is a big concern and several papers have reported on the monitoring of radioactive gas and airborne particles such as dust and aerosol [1)]. In contrast, not many data have been made available to the public regarding fallout in rain [2)]. The importance of direct measurements of fallout in rain is illustrated by the fact that the increase in the air dose rate was clearly correlated with rain-fall that occurred on March 21, as measured in Chiba city by the Japan Chemical Analysis Center. Diffusion simulation also showed that the radioactive plume arrived in the Kanto- area (Tokyo, Chiba, etc.) during this period of time. On the next day (March 22), radiation levels in the region's tap water spiked to their highest number (210 Bq/L for ^{131}I). This level is more than twice the recommended limit of 100 Bq/L for infants (although it is lower than the recommended limit of 300 Bq/L for adults), and consequently concerns about safety spread. In this respects, we monitored fallout in rain in Hiroshima from March to May, 2011 and we also checked several other sites, including Tokyo, Chiba, Fukushima, Osaka,

and Nara Prefectures.

2. Experimental

We began sampling rain-fall from March 20 to May 23, 2011, at the Graduate School of Engineering, in Hiroshima University, which is located in Higashi-Hiroshima city (34°40′N, 132°71′E). The school is 820.9 km from the Fukushima I Nuclear Power Plant (37°45′N, 141°27′E), about 40 km east of Hiroshima-city, Japan. Samples were collected by placing baskets on the roof of one of the lecture room buidlings during a heavy rain (the sampling volume was slightly more than about 1 L). During periods of lighter rainfall, samples were collected in a large area of roof gutter. Rain samples were also collected in several cities: Kashiwa city (Hananoi), Chiba Prefecture (35°90′N, 139°96′E) on March 21, April 11, and May 12; Nerima city (Shakujii), Tokyo (35°73′N, 139°58′E) on March 21 and April 11; Fukushima city (Mikawaminami), Fukushima (37°76′N, 140°46′E) on April 19; Hirano city (Hiranohonmachi), Osaka (34°62′N, 135°55′E) on April 8; Kitakatsuragi-gun (Koryo town), Nara (34°55′N, 135°72′E) on April 9. The distances of these locations from the Fukushima I plant are 194.7, 227.1, 63.2, 584.2, and 575.2 km, respectively. Collection periods varied from a few tens of minutes to approximately one day.

Following the collection in Hiroshima, the rainwater samples were chemically enriched prior to gamma-ray counting. Each rainwater sample, with volumes ranging from 0.030 to 1 L was poured into a stainless-steel tray, the surface of which was covered by a teflon sheet. First, 2 ml of CsCl (1 mol/L) was added to coprecipitate for iodine. Second, 2 ml of NaI (0.1 mol/L) was poured in to prevent volatilization of the iodine. Third, 0.15 ml of laundry starch was introduced to prevent residue scatter. Then, the sample was evaporated to dryness on an electric hot plate from 125 °C to 80° C, to 50 °C, at which point the dried residue was attached to a teflon sheet. Finally, the teflon sheet was encapsulated in a polystyrene petri dish (with a diameter of 5.2 cm) for gamma-ray counting.

All the rainwater samples were measured using our low-background high-purity germanium (HPGe) spectrometer in Higashi-Hiroshima. The spectrometer is a coaxial-type HPGe detector (EG & ORTEC, GEM-50195-P: 294 cc) with 12.5 cm lead, 5cm steel, 5 mm copper shields (LBR-2-2, SEIKO EG & G), and 5 mm acrylic for hermetic shielding to protect against radon in the air. The spectrometer is located at the Isotope Center, Hiroshima University. It is noted that the steel is special low-background steel, that was salvaged from the battle ship ``Mutsu."

Raw γ-ray energy spectra from 0.05 to 3.00 MeV were collected over the course of 80000 s (about one day) for each sample. The detector was calibrated using its own natural radioactive background. The detection efficiency for γ-rays has been measured with standard sources of known decay rates such as ^{137}Cs, ^{54}Mn, ^{88}Y, ^{60}Co, ^{109}Cd, ^{139}Ce, ^{51}Cr, and ^{85}Sr covering the energy range of 0.08 — 2.00 MeV, and it was compared with the GEANT4 simulation by taking each geometry into account. The details of the analysis are described elsewhere [3)].

3. Results

The first rainwater sample was collected on March 20, 2011 in Hiroshima. This sample showed no evidence of fission fragment γ-rays and all visible γ-ray peaks could be attributed to known background sources from the decays of the progeny of ^{238}U, ^{232}Th, and ^{40}K. These sources do not originate from the water, rather they are a part of the natural background present in our laboratory environment, including the impurities from the detector system. The first sample that showed fission products was collected on March 27 in Hiroshima, and the samples taken on subsequent days showed clear peaks attributable to ^{131}I at 364.5 keV, etc., ^{134}Cs at 604.7 keV, etc., and ^{137}Cs at 661.7 keV. Background spectrum was measured without a sample and a similar spectrum was also obtained by measuring a 1 L sample of Higashi-Hiroshima tap water. From this spectrum, we set a 3-sigma upper limit on the ^{131}I, ^{132}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{132}Te, and ^{7}Be activity in this sample to be 0.04, 0.03, 0.03, 0.03, 0.03, 0.03, and 0.27 Bq/L, respectively.

Several samples of rainwater collected in Chiba (Kashiwa) on March 21, April 11, and May 12, Tokyo (Nerima) on March 21 and April 11, Osaka (Hirano) on April 8, Nara (Kitakatsuragi) on April 9, and Fukushima (Fukushima) on April 19 were also checked for γ radioactivity using our spectrometer, and the samples were compared. Among these samples, the one collected in Kashiwa city, Chiba Prefecture on March 21 showed

Table 1 Fallout of radionuclides, in Bq/m^2, in rainwater via a rain fall event, March – May, 2011. Statistical uncertainties should be referred to ref. 3.

Date	Place	Rain (mm)	^{131}I	^{132}I	^{134}Cs	^{136}Cs	^{137}Cs	^{132}Te	^{7}Be
March 20	Hiroshima	7.5	ND	ND	ND	ND	ND	ND	ND
March 20 – 21	Chiba	23.5	140000	15000	15000	2300	18000	18000	2400
March 21 – 22	Tokyo	33	20000	3200	720	110	900	3800	160
March 27	Hiroshima	0.5	0.19	ND	0.075	ND	0.077	ND	4.9
April 8	Hiroshima	21	9.2	ND	ND	ND	ND	ND	39
April 8	Osaka	16.5	ND	ND	1.2	ND	0.92	ND	5.3
April 8 – 9	Nara	20.5	ND	ND	ND	ND	ND	ND	30
April 11	Chiba	2.5	25.5	ND	49	2.7	58	ND	7.6
April 11	Chiba	2.5	180	ND	170	8.1	220	ND	29
April 11	Tokyo	15.5	32	ND	110	5.1	130	ND	31
April 18	Hiroshima	4.5	ND	ND	0.68	ND	0.78	ND	34
April 19	Fukushima	18.5	550	ND	1400	45	1900	ND	300
April 22	Hiroshima	2.5	ND	ND	0.09	ND	0.06	ND	11
April 27 – 28	Hiroshima	12	ND	ND	ND	ND	ND	ND	15

the highest activity concentrations of 6072±51, 632±4, 766±3, 637±1, 97.6±0.8 and 752±2 Bq/L for ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs, ^{136}Cs and ^{137}Cs, respectively [3], which correspond to fallout/deposition of 140000, 15000, 15000, 2300, 18000, and 18000 Bq/m^2, respectively in Table 1. The fallout was calculated by the concentration multiplied by its precipitation for each day. The precipitation data was obtained by Hiroshima University Meteorological Dara Acquisition Data System (HIRODAS) [4] and the JMA(Japan Meteorological Agency)'s AMeDAS(Automated Meteorological Data Acquition System) for Hiroshima and the other places (Nerima, Abiko, Fukushima, Osaka, Katsuragi station), respectively.

4. Discussion

^{131}I was observed only on the early dates of March 27 and April 8, while ^{137}Cs and ^{134}Cs were observed on March 27, April 18, and April 22 in Hiroshima. This means that the radioactive cloud was initially richer in ^{131}I, as iodine is a more volatile element than cesium. The ^{131}I, ^{137}Cs, and ^{134}Cs activity concentrations in rainwater collected in Hiroshima reached 0.44±0.09 Bq/L on April 8, 0.17±0.01 Bq/L on April 18, and 0.15±0.01 Bq/L on April 18, respectively. A comparison between the highest activity level in Hiroshima and the results reported from other locations in the northern hemisphere is given elsewhere [3].

Our measured activity can be compared to the regulation set by the Ministry of Health, Labour and Welfare, Japan, which states that the maximum allowable levels of ^{131}I and ^{137}Cs in drinking water are 300 Bq/L (100 Bq/L for infants) and 200 Bq/L, respectively. The activity of 614±1 for ^{131}I collected in Nerima, Tokyo on March 21, and that of 6072±1 and 752±2 for ^{131}I and ^{137}Cs, respectively, collected in Kashiwa, Chiba on the same date, were beyond this regulation and coincided with the situation in which the activity of tap water at the water purification plant in Tokyo and Chiba exceeded this regulation on March 22. The maximum level of ^{131}I collected in Kashiwa city, Chiba, exceeded the IAEA Operational Intervention level for nuclear emergencies of 3000 Bq/L, as well. Other than these two cases in Chiba and Tokyo on March 21, all of the other results were below the regulation. It is noted that the observed reduction factors of about 10^{-5} and 10^{-4} after 52.6 days from March 21 to May 12 for the activity of ^{131}I and ^{137}Cs, respectively, are much larger than the expected decrease of about 10^{-2} and 0.997 because of the half-life of 8.0 days and 30.1 years, respectively. This is attributable to the fact that the activity largely depends on transport/deposition with rain. If a person were to drink 2 L of water per day containing the highest activity level of

Table 2 Comparison of fallout, in Bq/m^2, in rain that was measured in Hiroshima due to the Fukushima and Chernobyl reactor accidents [7] and the Hiroshima atomic bomb [8].

	^{131}I	^{134}Cs	^{137}Cs
Fukushima	9.4±0.4	0.85 ± 0.12	0.92 ± 0.14
Chernobyl	740	N.A.	N.A.
A-bomb	N.A.	N.A.	670±60

^{131}I that was measured in Chiba on March 21, then in one year he or she would receive a whole body dose of ~ 70 mSv, based on the procedure set by MEXT [5]. This can be compared with the ICRP (NRC) limits stating that no worker should exceed 50 mSv to the whole body or 500 mSv to the thyroid, and that no member of the public should exceed 1 mSv to the whole body. However, as described in the previous sentence, this is not the case.

Note that no fission products were detected in Nara, even though samples were collected on the same date and the distance between Osaka and Nara is only about 9 km. It has been suggested that mountain ranges provided western Japan with shelter from nonvolatile elements such as Cs, which was absorbed into aerosols [6]. A similar condition may have occurred in Nara, i.e., the transport of particulate material and radioactive plume may have been blocked by the Ikoma mountain (642 m height) between Osaka and Nara, which forms the Nara basin.

The comparison of fallout/deposition in rain that was measured in Higashi-Hiroshima due to the Fukushima and Chernobyl reactor accidents and the Hiroshima atomic bomb was listed in Table 2. It is noted that on the case for Chernobyl, since the only activity concentration was given in the paper [7], the same procedure as described in the previous chapter to obtain the fallout by multiplied with its precipitation via AMeDAS was conducted. Our obtained values measured at Higashi-Hiroshima, 40 km east to Hiroshima-city, due to the Fukushima accident can be also compared with the measurements in Hiroshima-city [9] and Nagoya (Y. Ogata in ref. 2), which were ND and 40±2 Bq/m^2, 4.3 and 8.4±0.9 Bq/m^2, and 4.1 and 7.7 ± 0.8 Bq/m^2 for ^{131}I, ^{134}Cs, and ^{137}Cs, respectively. It is also noted that the deposition of ^{137}Cs from global fallout prior to 1980 was about 3700 Bq/m^2 [8].

5. Conclusion

Fallout in rain due to the Fukushima reactor accident was monitored for about two months (from March 20 to May 23, 2011) in Higashi-Hiroshima city, Hiroshima. The activities of nuclear fission products reached 0.44± 0.09 Bq/L on April 8 and 0.17±0.01 and 0.15±0.01 Bq/L on April 18 for ^{131}I, ^{137}Cs, and ^{134}Cs, respectively. Several rainwater samples collected in Chiba (Kashiwa), Tokyo (Nerina), Osaka, Nara, and Fukushima were also checked, and the Chiba data showed the highest activities of 6072±1, 752±2, and 637±1 Bq/L on March 21 for ^{131}I, ^{137}Cs, and ^{134}Cs, respectively. The activity of 614±1 for ^{131}I measured in Nerima, Tokyo on March 21 was also beyond the regulation. Other than these two cases from Chiba and Tokyo on March 21, all of the other results were below the regulation. Fortunately, the observed reduction of these activities after about 50 days was much larger than the expected decrease due to radioactive decay. Our measured fallout/deposition in rain due to the Fukushima reactor accident was also compared with that due to the Chernobyl reactor accident and the result in black rain caused by fallout of the Hiroshima atomic bomb.

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References

1) T. Ohara, Y. Morino, A. Tanaka, “Atmospheric behavior of radioactive materials from Fukushima Daiichi Nuclear Power Plant”, *J. Natl. Inst. Public Health*, 60 (4), 292, (2011)(in Japanese); G. Brumfiel, “Fallout forensics hike raidation toll”, *Nature*, 478, 435 (2011): T. W. Bowyer, et al., “Elevated radioxenon detected remotely following the Fukushima nuclear accident”, *J. Environ. Radioact.* 102, 681 (2011): L. E. Sinclair, et al., “Aerial measurement of radioxenon concentration off the west coast of Vancouver Island following the Fukushima reactor accident”, *J. Environ. Radioact.* 102, 1018 (2011): J. Diaz Leon, et al., “Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle WA, USA”, *J. Environ. Radioact.* 102, 1032 (2011): M. Manolopoulou, et al., “Radioiodine and

radiocesium in Thessaloniki, Northern Greece due to the Fukushima nuclear accident", *J. Environ. Radioact.* 102, 796 (2011); "Radiation measurements and radioecological aspects of fallout from the Fukushima nuclear accident", *J. Radioanal. Nucl. Chem.*, DOI 10.1007/s10967-011-1386-2 (2011): K. Fushimi, et al., "Measurement of airborne radioactivity from the Fukushima reactor accident in Tokushima, Japan", arXiv: 1104.3611[nucl-ex]: K. Shizuma, "Survey of radioactivity concentration in the Kagamiyama area's atmosphere, Hiroshima University Homepage: http://www.hiroshima-u.ac.jp/en/top/news_events/earthquake/sokuteikekka/: S. MacMullin, et al., "Measurement of airborne fission products in Chapel Hill, NC, USA from the Fukushima Dai-ichi reactor accident", *J. Environ. Radioact.* 112, 165 (2012).

2) E. Norman, et al., "Observations of Fallout from the Fukushima Reactor Accident in San Francisco Bay Area Rainwater", arXiv: 1103.5954[nucl-ex]: Y. Ogata, "Fallout from Fukushima accident in Nagoya", *Proceedings of the Seminar at Nagoya by Radiation Education Forum*, January, 5 (2012) (in Japanese): "Radioiodine and radiocesium in fallout with rain in Nagoya from March to May 2011", *Proceedings of the Annual meeting of Japan Health Physics Society*, October, 102 (2011) (in Japanese): The 7th International Workshop on Ionizing Radiation Monitoring, December (2011), to be published in *Radiation Measurements* (2012): D. Pittauerova, B. Hettwig, H. W. Fischer, "Fukushima fallout in Northwest German environmental media", *J. Environ. Radioact.* 102, 877 (2011): B. T. Cleveland, et al., "Activities of γ -ray emitting isotopes in rainwater from Greater Sudbury, Canada following the Fukushima incident", *Can. J. Phys.*, 90, 599 (2012)

3) R. Hazama and A. Matsushima, "Measurement of fallout with rain from Fukushima Reactor Accident in Hiroshima and several sites in Japan by HPGe spectrometer"to be published in *J. Radioanal. Nucl. Chem.*

4) Hiroshima University Meteorological Dara Acquisition Data System (HIRODAS) : http://home.hiroshima-u.ac.jp/hirodas/

5) Dose conversion coefficient of 1.63×10^{-8} Sv/Bq has been applied: http://www.mext.go.jp/component/b_menu/shingi/giji/_icsFiles/afieldfile/2012/01/13/1315036_5.pdf

6) N. Kinoshita, et al., "Assessment of individual radionuclide distributions from Fukushima nuclear accident covering central-east Japan", *PNAS*, 108, 19526 (2011): T. J. Yasunari, et al., "Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident", *PNAS*, 108, 19530 (2011).

7) K. Shizuma, et al., "Observation of fallout in Hiroshima caused by the reactor accident at Chernobyl", *Int. J. Radiat. Biol.*, 51, 201, (1987).

8) K. Shizuma, S. Endo, Y. Fujikawa, "Isotope Ratios of $^{235}U/^{238}U$ and $^{137}Cs/^{235}U$ in Black Rain Streaks on Plaster Wall Caused by Fallout of the Hiroshima Atomic Bomb", *Health Physics*, 102, 154, (2012).

9) Monitoring info of environmental radioactivity level by the MEXT: http://radioactivity.mext.go.jp/old/ja/monitoring_by_prefecture_fallout/