

Measurement of fallout with rain in Hiroshima and several sites in Japan from the Fukushima reactor accident

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Abstract Fallout with rain from the Fukushima reactor accident was monitored for about two months in Higashi-Hiroshima City, Hiroshima, Japan, from March 20 to May 23, 2011. Gamma-ray (γ -ray) spectra measured using a low background HPGe spectrometer showed clear evidence of fission products— ^{131}I , ^{137}Cs , and ^{134}Cs . The ^{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 ± 0.09 on April 8, 0.17 ± 0.01 on April 18, and 0.15 ± 0.01 Bq/L on April 18, 2011, respectively. These activity levels were compared with global results collected from the Northern Hemisphere. Several samples of rainwater that were 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 by our spectrometer and compared. Among these samples of rainwater collected at different locations in Japan, the one collected at Kashiwa City, Chiba Prefecture, on March

21 showed the highest activity concentrations of 6072 ± 1 , 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. The health risks due to these high activity levels were considered in terms of several regulations. The ratio of the activities for the isotopes ^{131}I to ^{137}Cs and ^{134}Cs to ^{137}Cs were compared with the other measurements and discussed.

Keywords Radioactive fallout · Fukushima nuclear accident · Fission product radionuclides · γ -ray spectrometry

Introduction

The big earthquake (M9.0) at 14:46 p.m. on March 11, 2011 was followed by a big tsunami that caused the severe accident at Fukushima Daiichi Nuclear Power Plant. Due to the blackout, hydrogen detonation occurred at Unit-1 at 15:36 p.m. on March 12, Unit-3 at 11:01 a.m. on March 14, and explosion occurred at Unit-2 at around 6 a.m. on March 15, sequentially, and the estimated amounts of 160, 18, and 15×10^{15} Bq were released for ^{131}I , ^{134}Cs , and ^{137}Cs , respectively [1, 2]. Its spread/distribution of radioactivity was a big concern, and monitoring radioactive gas and airborne particles such as dust and aerosol were reported in several papers [3–9]. However, not much data on fallout with rain [6, 10–13] has been open to the public, compared with that in the air. The importance of direct measurement of fallout with rain is pointed out by the fact that the rise of air dose rate was clearly correlated with rainfall on March 21, measured at Chiba City by the Japan Chemical Analysis Center [14]. Diffusion simulation also showed that radioactive plume was delivered to the Kanto area during this time period [15–17]. On the next day

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(March 22), radiation levels spiked with the highest number of 210 Bq/L for ^{131}I in Kanto area (Tokyo, Chiba, etc.) tap water [18], and concerns about safety spread since there was more than twice the recommended limit of 100 Bq/L for infants (lower than that of 300 Bq/L for adults). In these respects, we monitored fallout with rain in Hiroshima from March to May 2011, and also checked several sites/dates including the above mentioned Tokyo, Chiba, and Fukushima, Osaka, Nara Prefecture.

Experimental

We started sampling rainfall at the Graduate School of Engineering, Hiroshima University, located at Higashi-Hiroshima City (34°40'N, 132°71'E), where the distance is 820.9 km from the Fukushima Daiichi Nuclear Power Plant (37°45'N, 141°27'E), about 40 km east of Hiroshima-City, Japan, from March 20 to May 23, 2011. Samples were collected directly by placing baskets on the roof of a lecture room building during a heavy rainfall (sampling volume > about one liter) and by a large area of roof gutters in case a little rainfall. Rain samples were also collected in several cities at 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; and Kitakatsuragi-gun (Koryo Town), Nara (34°55'N, 135°72'E) on April 9, 2011. Distances from the Fukushima

Daiichi Plant were 194.7 km, 227.5 km, 63.2 km, 584.2 km, and 575.2 km, respectively (Fig. 1). Collection periods varied from a few tens of minutes to approximately one day.

Following collection, chemical processing of enrichment was done to the rainwater samples in Hiroshima before gamma-ray counting. Rainwater samples varied in volume from 0.030 to 1 liter and were poured into a stainless tray whose surface was covered by a Teflon sheet. First, 2 mL of CsCl (1 mol/L) was added to a co-precipitate for iodine. Second, 2 mL of NaI (0.1 mol/L) was poured to prevent the volatilization of 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 to 80, and at 50 °C, dried residue was attached on a Teflon sheet. In the end, 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 by our low-background, high-purity germanium (HPGe) spectrometer in Higashi-Hiroshima (see Table 1). The spectrometer was a coaxial-type HPGe detector with 12.5 cm of lead, 5 cm of steel, 5 mm of copper shield (LBR-2-2, SEIKO EG&G), and 5 mm of acrylic for hermetic shielding to protect against radon in the air, and was located at the Isotope Center at Hiroshima University [20]. It should be noted that this steel was special low-background steel salvaged from the battle ship “Mutsu.” The contamination of ^{60}Co in steel was checked to be 0.03 ± 0.004 (Bg/kg) at the maximum for Mutsu [19] and 0.37 Bq/kg for modern steel [20].

The raw γ -ray energy spectra from 0.05–3.00 MeV were collected for the live time of 80000 sec (about one day) for

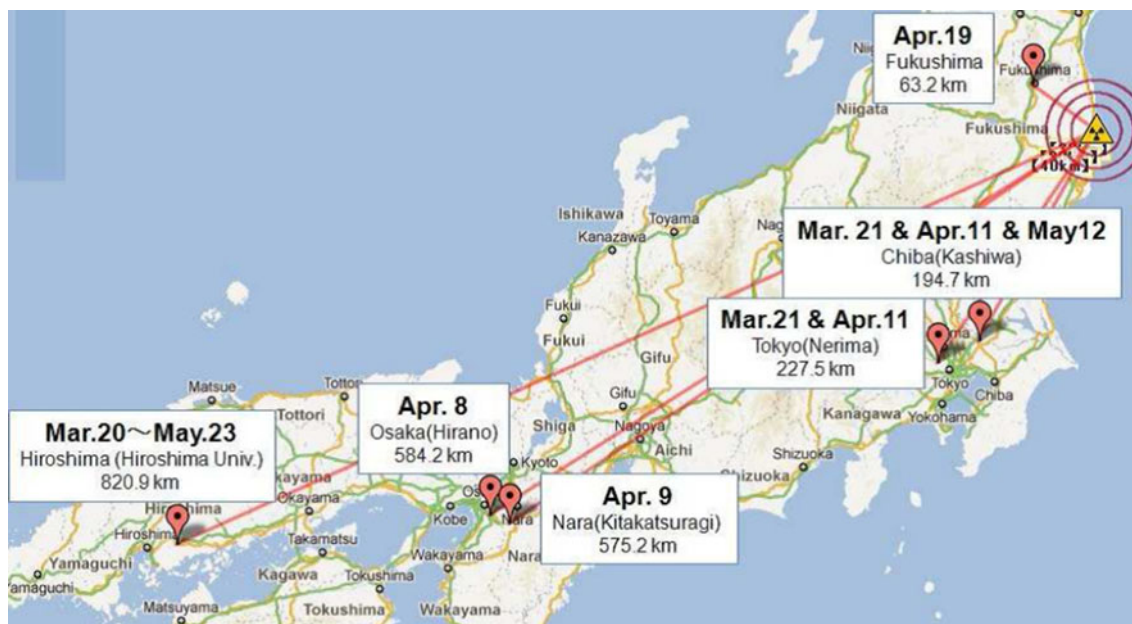


Fig. 1 Map of sampling site

The sampling dates and measured rainwater activities for the first 73 days of data are listed in Table 2. We cross-checked for consistency between the activities obtained from different branches of the same isotope. As they were in agreement and the systematic error dominates, we only gave the value for the strongest branch. ^7Be activity was also compared since ^7Be is recognized as an atmospheric tracer due to the fact that its measurement gives information about diffusion properties of the high troposphere and low stratosphere, as well as a description of environmental processes such as precipitation, washout, atmospheric particle deposition, and depositional patterns of airborne contaminants [22].

Several samples of rainwater were 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, and were also checked for their γ radioactivity by our spectrometer and compared. Among these samples of rainwater collected at different locations in Japan, the one collected at Kashiwa City, Chiba Prefecture, on March 21 showed the highest activity concentrations of 6072 ± 1 , 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. We believe the activity concentration in Fukushima would have been higher if we collected the rainwater samples in March, not around the middle of April, 2011, and we regret that we were unable to do so.

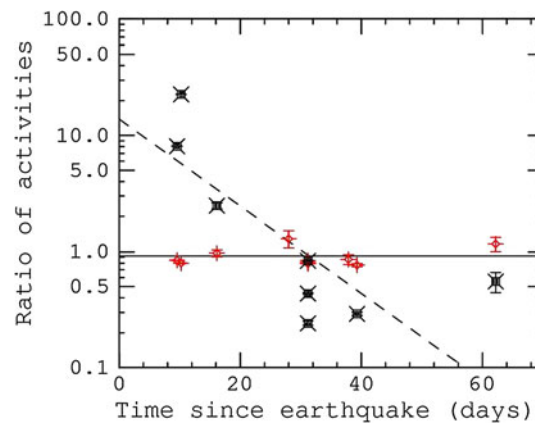


Fig. 3 Ratio of activities for the isotopes ^{131}I to ^{137}Cs (black) and ^{134}Cs to ^{137}Cs (red). The error bar shows the statistical error only as the systematic errors cancel to a large degree in the ratios. The fits to the ratios as a function of time were performed with fixed decay time constants (dashed line). The weighted average ratio of ^{134}Cs to ^{137}Cs was found to be 0.92 ± 0.03 , which was plotted as a black line

Using all of our measured data in Table 2, the ratio of activities of ^{131}I and ^{134}Cs to ^{137}Cs were calculated and plotted against the time since the earthquake (days) in Fig 3. The weighted average ratio of ^{134}Cs to ^{137}Cs was found to be 0.92 ± 0.03 in agreement with the value of $0.8 - 0.9$, reported in the whole investigated region, including Fukushima Prefecture [23]. This equally produced ^{134}Cs will be an indication of the nuclear reactor accident rather than a

Table 2 Activity concentrations, in Bq/L, of radionuclides in rainwater via rain fall events, March–May, 2011

Date of Sampling	Place	^{131}I	^{132}I	^{134}Cs	^{136}Cs	^{137}Cs	^{132}Te	^7Be
March 20	Hiroshima	ND	ND	ND	ND	ND	ND	ND
March 20–21	Chiba	6072 ± 1	632 ± 4	637 ± 1	97.6 ± 0.8	752 ± 2	766 ± 3	100 ± 2
March 21–22	Tokyo	614 ± 1	98.2 ± 0.9	21.7 ± 0.2	3.33 ± 0.09	27.2 ± 0.2	115 ± 1	4.73 ± 0.22
March 27	Hiroshima	0.38 ± 0.01	ND	0.149 ± 0.007	ND	0.153 ± 0.008	ND	9.73 ± 0.16
April 8	Hiroshima	0.44 ± 0.09	ND	ND	ND	ND	ND	1.84 ± 0.09
April 8	Osaka	ND	ND	0.073 ± 0.008	ND	0.056 ± 0.008	ND	0.32 ± 0.05
April 8–9	Nara	ND	ND	ND	ND	ND	ND	1.47 ± 0.09
April 11	Chiba	10.2 ± 0.1	ND	19.6 ± 0.2	1.07 ± 0.04	23.3 ± 0.2	ND	3.05 ± 0.16
April 11	Chiba	73.1 ± 0.3	ND	69.0 ± 0.2	3.24 ± 0.07	87.4 ± 0.3	ND	11.6 ± 0.3
April 11	Tokyo	12.6 ± 0.5	ND	42.0 ± 0.2	2.05 ± 0.15	52.1 ± 0.3	ND	12.3 ± 0.4
April 18	Hiroshima	ND	ND	0.15 ± 0.01	ND	0.173 ± 0.012	ND	7.48 ± 0.25
April 19	Fukushima	29.6 ± 0.4	ND	77.6 ± 0.2	2.43 ± 0.09	101 ± 3	ND	16.2 ± 0.3
April 22	Hiroshima	ND	ND	0.036 ± 0.004	ND	0.025 ± 0.003	ND	4.29 ± 0.13
April 27–28	Hiroshima	ND	ND	ND	ND	ND	ND	1.27 ± 0.10
May 7	Hiroshima	ND	ND	ND	ND	ND	ND	2.29 ± 0.12
May 10	Hiroshima	ND	ND	ND	ND	ND	ND	0.30 ± 0.03
May 12	Chiba	0.036 ± 0.006	ND	0.076 ± 0.007	ND	0.065 ± 0.007	ND	ND
May 23	Hiroshima	ND	ND	ND	ND	ND	ND	0.60 ± 0.04

Statistical uncertainties are $\pm 1 \sigma$

Table 3 The highest activities (Bq/L) of radioiodine and radiocesium and their detected dates in *parentheses* (month, day) in the rainwater samples, reported in the world continuous surveys, except for occasional spot-checks collected at Chiba, Tokyo, Fukushima, Osaka, and Nara

Place	¹³¹ I	¹³⁴ Cs	¹³⁷ Cs
Hiroshima, Japan	0.44 ± 0.09 (4.8)	0.15 ± 0.01 (4.18)	0.17 ± 0.01 (4.18)
Nagoya, Japan [11]	1.4 ± 0.1 (4.8)	0.28 ± 0.02 (4.18)	0.24 ± 0.02 (4.18)
San Francisco, USA [10]	19.8 ± 2.0 (3.24)	0.55 ± 0.05 (3.24)	0.65 ± 0.06 (3.24)
Thessaloniki, Greece [6]	0.69 ± 0.18 (3.28–29)	ND	ND
Bremen, Seefeld, Germany [12]	0.43 ± 0.03 (3.20–4.10)	ND	ND
Sudbury, Canada [13]	0.668 ± 0.044 (4.3)	0.0168 ± 0.029 (4.16)	0.0277 ± 0.049 (4.16)

nuclear weapon test since the formation of ¹³⁴Cs via neutron capture on ¹³³Cs requires a length of criticality. Even though iodine and cesium have different chemical properties, the ratio of ¹³¹I to ¹³⁷Cs can be used to obtain information about when the nuclear reactions terminated. This iodine–cesium dating was applied to the data of the Tokyo Electric Power Company (TEPCO) for seawater samples at several monitoring posts near the troubled reactor and a water sample taken from the spent-fuel cooling pool of the Unit-4 reactor building by Matsui [24]. The same exponential function was applied to our current data:

$$\frac{I_{\text{activity}}}{Cs_{\text{activity}}}(t) = \frac{f_I}{f_{Cs}} \frac{\tau_{Cs}}{\Delta t \cdot \ln(2)} \left(\frac{1}{2}\right)^{t/\tau_I} \quad (2)$$

where f_I and f_{Cs} are the fractions of ¹³¹I and ¹³⁷Cs per fission (2.89×10^{-2} and 6.19×10^{-2} , respectively; [25]) τ_I and τ_{Cs} are the lifetimes of ¹³¹I and ¹³⁷Cs; and Δt is the length of time the reactor has been active. The best fit value for Δt was found to be 1.5 years with large deviations ($\chi^2/\text{dof} \geq 5$), but are in good agreement with the one year reported by Matsui.

Discussion

The ¹³¹I was observed only at the early dates on March 27 and April 8, while ¹³⁷Cs and ¹³⁴Cs were observed on March 27, April 18, and April 22 in Hiroshima. This means that the radioactive cloud was first richer in ¹³¹I, as iodine is a more volatile element than cesium. The ¹³¹I, ¹³⁷Cs, and ¹³⁴Cs activity concentrations in rainwater collected in Hiroshima reached 0.44 ± 0.09 on April 8, 0.17 ± 0.01 on April 18, and 0.15 ± 0.01 Bq/L on April 18, 2011, respectively. The highest activity level in Hiroshima was compared with global results (Table 3), which were collected in the Northern Hemisphere. Among these results on continuous surveys, except for occasional spot checks such as the results in Chiba, Tokyo; Fukushima, Osaka; and Nara, the highest values for all radioiodine and radiocesium were measured at Berkeley, Oakland, and Albany, California, USA. Note that all the dates of the highest activities reported

in Western countries were earlier than that of western Japan (Hiroshima and Nagoya). This will be affected by specific local weather conditions on these dates, and will undoubtedly be a complicated function of many variables including release rates at the Fukushima reactor site and wind patterns and velocities. These may be attributed to the general trend of typical winter pressure patterns in Japan, such as the high-pressure area to the west and low-pressure area to the east, and the Westerlies (jet stream flowing west to east), which are strongest in the winter hemisphere.

This maximum activity can be compared to the regulation set by the Ministry of Health, Labour, and Welfare, Japan, on ¹³¹I and ¹³⁷Cs allowed in drinking water of 300 Bq/L (100 Bq/L for infants) and 200 Bq/L, respectively¹. The activity collected at Nerima, Tokyo, on March 21 of 614 ± 1 for ¹³¹I was also beyond this regulation and coincided with the situation that the activity of tap water at the water purification plant in Tokyo and Chiba exceeded this regulation on March 22. The maximum level of ¹³¹I collected at Kashiwa city, Chiba, exceeded the IAEA Operational Intervention level for nuclear emergencies at 3000 Bq/L, too. Except for the two cases in Chiba and Tokyo on March 21, all 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 its activity of ¹³¹I and ¹³⁷Cs, respectively, are much larger than the expected decrease of about 10^{-2} and 0.997 due to the half-life of 8.0 days and 30.1 years, respectively. This is attributable to the fact that the activity largely depends on its transport/deposition with rain. If a person were to drink two liters of water per day containing the highest activity level of ¹³¹I in Chiba on March 21, then in one year he or she would receive a whole body dose of about 70 mSv, based on the procedure set by the MEXT [26]. This can be compared with the ICRP (NRC) limits that no worker exceeds 50 mSv to the whole body or 500 mSv to the thyroid, and that no member of the public exceeds 1 mSv to the whole body, but as described in the previous sentence, this is not the case.

¹ Since April 2012, the Ministry has set this regulation back to being 10 Bq/L, which had been applied until March 16, 2011.

Note that no fission products were detected in Nara, even though the same date and the distance between Osaka and Nara is only about 9 km. It has been suggested that western Japan was sheltered by mountain ranges from nonvolatile elements such as Cs, which was absorbed into aerosols [23, 27]. 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.

Conclusion

Fallout with rain from 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 , 0.15 ± 0.01 on April 18 for ^{131}I , ^{137}Cs , and ^{134}Cs , respectively. Our result was compared with other reported data from the Northern Hemisphere, and these highest activities and their detected dates in rainwater samples will help to understand the release of radioactive fission products and their transport in the atmosphere. Several rainwater samples collected at Chiba (Kashiwa), Tokyo (Nerima), Osaka, Nara, and Fukushima were also checked. Chiba data showed the highest activities of 6072 ± 1 , 752 ± 2 , 637 ± 1 Bq/L on March 21, 2011 for ^{131}I , ^{137}Cs , and ^{134}Cs , respectively. The activity collected at Nerima, Tokyo, on March 21, 2011 of 614 ± 1 for ^{131}I , was also beyond the regulation. Except for the two cases in Chiba and Tokyo on March 21, 2011, all 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 its decay-out. However, regular monitoring would be advised. The ratio of the activities for the isotopes ^{131}I to ^{137}Cs and ^{134}Cs to ^{137}Cs were also checked and found to be consistent with the other measurements.

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