Early air sampling in Higashi-Hiroshima after the Fukushima Daiichi Nuclear Power Plant accident and subsequent sampling in Minami-Souma City from October 2011 to September 2012

Kiyoshi SHIZUMA*, Tsuyoshi KAJIMOTO, Satoru ENDO

Graduate School of Engineering, Hiroshima University, 1-4-1 Kagamiyama, Higashi-Hiroshima 739-8527, JAPAN
*shizuma@hiroshima-u.ac.jp

Soon after the accident that occurred at the Fukushima Daiichi Nuclear Power Plant on March 11, 2011, air sampling was performed in Higashi-Hiroshima (about 800 km from Fukushima). The sampling began on March 20, and a high-volume air sampler was used. The first radionuclide observed was ^{131}I (on March 30), and ^{137}Cs, ^{134}Cs, ^{136}Cs, and ^{132}Te were observed thereafter. According to the sampling, the maximum concentration in the air occurred on April 7 and again on April 18. After April 30, no radionuclides released due to the accident were observed. Since ^{137}Cs and ^{134}Cs have a long half-life (30 and 2 y, respectively), all of the air filters were measured again in February through March of 2012 using a well-type Ge detector. The results of these measurements showed that ^{137}Cs was already observed on March 21. Later on, air sampling was performed at Minami-Souma City, Fukushima (October 2011 to September 2012). The purpose of this sampling was to investigate whether any radionuclides were released from the forest and flew up into the air from the ground, or were carried by the wind in winter or via pollen in spring. Results showed that the radioactive concentration was quite low and no seasonal variation occurred, indicating that no radionuclides flew up into the air from the ground, nor were any released from the forest via wind or pollen.

Key Words : *early dust sampling, radionuclides, Hiroshima, fly up, Minami -Souma*

1. Introduction

The nuclear reactor accident that occurred on March 11, 2011 resulted in serious radioactive contamination[1)] in Fukushima. Fission products were released into the air during the period of March 15–21[2)], and the residents in the area suffered external and internal exposure to radionuclides. Kamada et al.[3)] used the urinary bioassay method to estimate external and internal exposure levels, including effective organ doses in the thyroid gland, for 15 residents in Iitate Village and Kawamata Town. Previously, we had measured radioactive fallout that was released in the air due to the reactor accident at Chernobyl[4)] and was transported northward via the global wind. With regard to the accident that occurred at the Fukushima Daiichi Nuclear Power Plant, it was thought that radioactive fallout might not be transported to western Japan because, unlike the Chernobyl accident, the reactor accident at Fukushima did not involve an explosion of the reactor core. However, as it turned out, the local wind blowing through Fukushima did transport radioactive plumes to western Japan. We began performing air sampling on March 20, and we continued until the end of May. Our results were compared with other work [5)].

In September 2011, we started an environmental radioactivity survey in Minami-Souma City in Fukushima. Residents were anxious about the radioactivity level in foods, and they also wanted to know if radionuclides would be released from the forest and be carried up into the air by the wind in winter and via pollen in spring. To clarify these uncertainties, air sampling was performed in Minami-Souma City from October 2011 to September 2012. The results are also shown in this work.

2. Materials and Methods

Air sampling began on March 20, 2011 on the campus of Hiroshima University at Higashi-Hiroshima, which is located about 800 km from Fukushima. The location of Higashi-Hiroshima is shown in Fig. 1. The sampling was performed using a potable-type high-volume air sampler (Sibata AN500H; Sibata Scientific Technology Ltd.) with a 500 l/min capacity and a 110-mm-diam glass fiber filter (GB100R; Advantec Co. Ltd.). A charcoal filter was not used, thus gaseous ^{131}I might not have been fully collected in this sampling.

The air sampler was placed outdoors 1 m above the ground under the roof of the radiation facility. To detect the starting time of the contamination of the air, the sampling was done during two separate time periods each day—three hours in the morning and three hours in the afternoon—until March 31. After April 1, sampling was done for 8 hours per day (from 9 a.m. to 17 p.m.; total volume was 240 m^3). The dust filter was folded into a 3 cm × 3 cm form and pressed into a plastic container. The container was placed on the top of the low-background well-type Ge detector [6] (Seiko EG&G, GWL 120230-S) for gamma-ray measurement.

Since both ^{137}Cs and ^{134}Cs have a long half-life (30 and 2 y, respectively), the dust filters were measured again from February through March of 2012. The same dust filter was folded into a (13-mm-diam)×(35-mm-length) form before being placed into the well port of the Ge detector.

Air sampling was also performed at Minami-Souma City, Fukushima from October 2011 to September 2012. Air sampling was carried out in two to six places per day every two to three months. Sampling time was 30 min for each place. All of the locations are shown in Fig. 2.

3. Results and Discussion

The first radionuclide observed was ^{131}I (on March 30). The maximum concentration in the air occurred on April 7 and then again on April 18. A typical gamma-ray spectrum is shown in Fig. 3.

The detected fission products were ^{131}I, ^{137}Cs, ^{134}Cs, ^{132}Te, and ^{136}Cs. Small amounts of ^{132}Te and ^{136}Cs were observed on April 7.

The time variation of radionuclide concentration in the air is shown in Fig. 4. The maximum concentration was observed on April 7, and the second maximum was observed on April 18. A revised variation of concentration for ^{137}Cs and ^{134}Cs was obtained from the re-measurement of the filters using the well-type Ge detector. The results are shown in Fig. 5.

Momoshima et al.[4] detected fission products from the Fukushima reactor accident that were transported to Fukuoka, Kyushu. They collected air dust using two high-volume air samplers: one was equipped with a quartz fiber filter, and the other was equipped with an activated charcoal bag to collect gaseous ^{131}I. They reported that the particle fraction was 30–67% of the total ^{131}I. Based upon their observations, the radionuclides observed, and their concentrations on April 6 , were ^{131}I (particle) at 5.07 mBq/m^3, ^{134}Cs at 4.04 mBq/m^3, and ^{137}Cs at 4.53 mBq/m^3. These results are in good agreement with our observations. However, their maximum appeared on April 6 for ^{131}I, while the maximum from our work appeared on April 7. The second maximum was clearer in Higashi-Hiroshima. Momoshima et al. reported the initial detection of ^{131}I on March 17, and it was detected every day from March 22 to April 23. The first observation of ^{131}I in the present work occurred on the afternoon of March 21.

Weather simulations done by the governments of

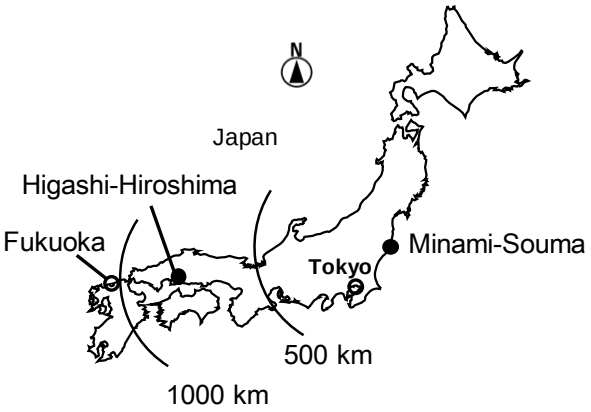

Fig. 1 Location of Higashi-Hiroshima and Minami-Souma.

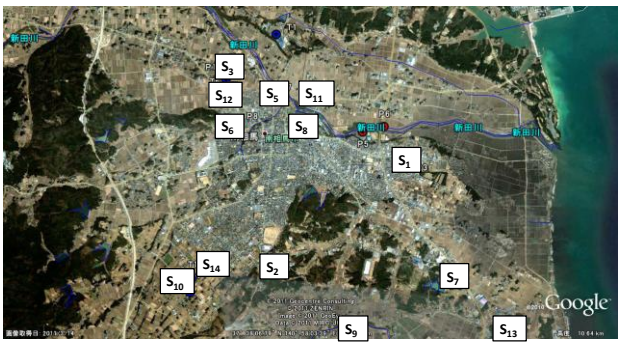

Fig. 2 Air sampling locations S_1 to S_{14} in Minami-Souma City. Air dust sampling was carried out in two to six places per day for every two to three months from October 2011 to September 2012.

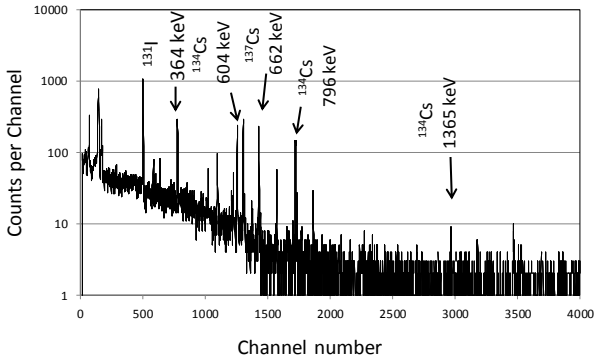

Fig. 3 An example of a gamma-ray spectrum measured on April 8, 2011.

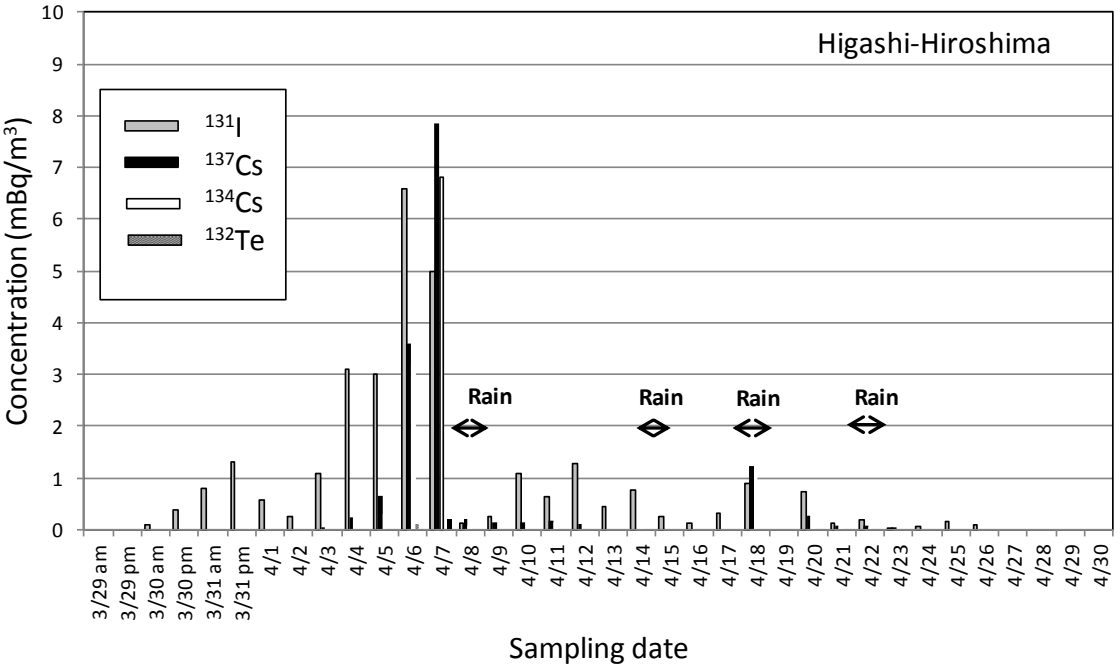

Fig. 4 Variation of radionuclide concentration in air detected at Higashi-Hiroshima.

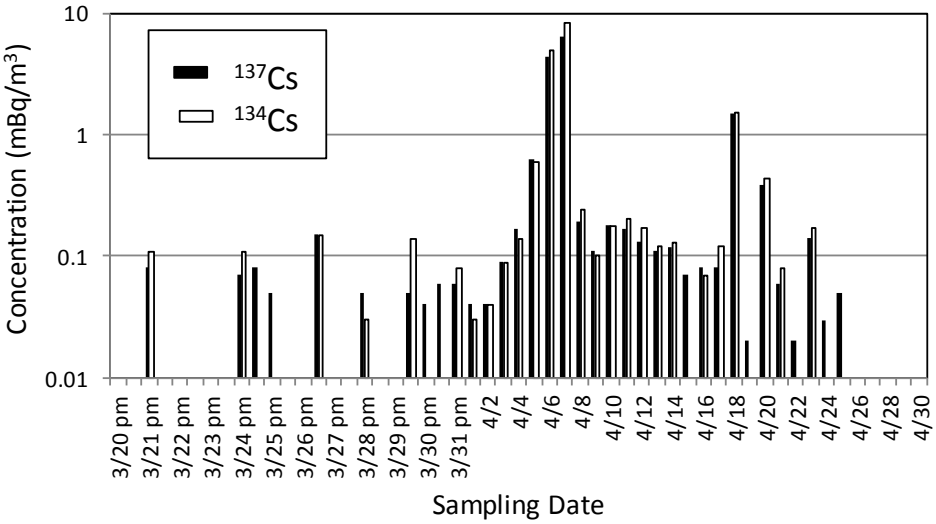

Fig. 5 Variation of ^{137}Cs and ^{134}Cs concentration in air.

Germany[7] and Japan[8] predicted the diffusion of a radioactive plume to the area west of Japan. The present results agree well with these simulations.

Air dust sampling was performed in Minami-Souma City from October 2011 to September 2012. Only ^{137}Cs and ^{134}Cs were detected, and at quite low levels. The results from two through six locations were averaged for each month, and they are shown in Table 1 and Fig. 6. The sum of ^{137}Cs and ^{134}Cs was in the range of 0.6–2.4 mBq/m^3, and no seasonal variation was observed. This indicates that the amount of radionuclides that flew up from the ground or were released from the forest was quite low. Among the residents of this region, the radiation doses that occurred through inhalation are negligible today.

4. Conclusion

Fission products that were released into the air after the Fukushima Daiichi Nuclear Power Plant accident on March 11, 2011, were detected in Higashi-Hiroshima, which is located 800 km from Fukushima. The maximum concentration in the air was observed on April 6–7, 2011, with levels of 6.6 mBq/m^3 for ^{131}I, 7.8 mBq/m^3 for ^{137}Cs, 6.8 mBq/m^3 for ^{134}Cs, 0.19 mBq/m^3 for ^{136}Cs, and 0.11 mBq/m^3 for ^{132}Te.

Table 1. Average concentration of ^{137}Cs and ^{134}Cs observed in Minami-Souma City from October 2011 to September 2012.

Month/Year	Location	^{137}Cs(mBq/m^3)	^{134}Cs(mBq/m^3)	^{137}Cs+^{134}Cs (mBq/m^3)
10/2011	S1, S2, S3, S4, S5, S6	1.0 ± 0.0	0.6 ± 0.1	1.6 ± 0.2
12/2011	S4, S5, S7	0.7 ± 0.1	0.3 ± 0.1	1.0 ± 0.2
3/2012	S1, S2, S8, S9, S10	1.0 ± 0.4	0.5 ± 0.3	1.5 ± 0.7
4/2012	S1, S2, S12, S13, S14	0.4 ± 0.4	0.3 ± 0.2	0.7 ± 0.6
5/2012	S1, S2	1.5 ± 0.8	0.9 ± 0.5	2.4 ± 1.3
7/2012	S1, S2	1.4 ± 0.2	0.6 ± 0.2	2.0 ± 0.3
9/2012	S1, S2	0.5 ± 0.1	0.1 ± 0.2	0.6 ± 0.3

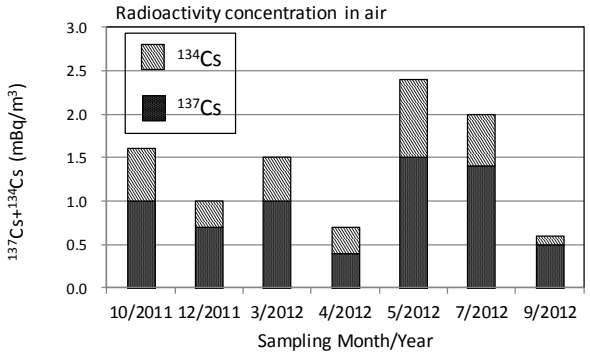

Fig. 6 Average concentration of ^{137}Cs and ^{134}Cs observed in Minami-Souma City from October 2011 to September 2012.

Dust sampling was performed in Minami-Souma City from October 2011 to September 2012 Results showed that the total radioactive concentrations of ^{137}Cs and ^{134}Cs were in the range 0.6–2.4 mBq/m^3, and no seasonal variations were
observed. This indicates that the amount of radionuclides that were carried up into the air by the wind is quite low.
In addition, with regard to radiation exposure through inhalation, the residents of the area displayed negligible effects.

Acknowledgement

The authors sincerely thank Mr. Yuji Sakurai in Minami-Souma City for his cooperation throughout the sampling.

References

1) Monitoring information of environmental radioactivity level about aircraft monitoring reported 28.09.2012. http://radioactivity.mext.go.jp/ja/ (accessed 05.11.2012).
2) K. Nagaoka, S. Sato, S. Araki, U. Ohta, Y. Ikauchi, Changes of radionuclides in the environment in Chiba, Japan after the Fukushima nuclear power plant accident, Health Phys. 102(4) (2012) 437-442.
3) N. Kamada, O. Saito, S. Endo, A. Kimura, K. Shizuma, Radiation doses among residents living 37 km northwest of the Fukushima Dai-ichi nuclear
4) power plant, J. Environ. Radioactiv. 110 (2012) 84-89.
5) K. Shizuma, K. Iwatani, H. Hasai et al., Observation of fallout in Hiroshima caused by the reactor accident at Chernobyl, Int. J. Radiat. Biol. 51(2) (1987) 201-207.
6) N. Momoshima, S. Sugihara, R. Ichikawa, H. Yokoyama, Atmospheric radionuclides transported to Fukuoka, Japan remote from Fukushima Dai-ichi nuclear power plant complex following the nuclear accident. J. Environ. Radioactiv. (2011), doi:10.1016/j.jenvrad.2011.09.001.
7) K. Shizuma, K. Fukami, K. Iwatani, H. Hasai, Low-background shielding of Ge detectors for the measurement of residual ^{152}Eu radioactivity induced by neutrons from the Hiroshima atomic bomb, Nucl. Inst. Meth. B66 (1992) 459-464.
8) http://www.dwd.de/bvbw/appmanager/bvbw/dwdwwwDesktop?_nfpb=true&_pageLabel=dwdwww_start&_nfls=false (accessed 04.20.2012).
9) Japan Meteorological Agency Global Tracer Transport Model, Ministry of Education Culture, Sports, Science& Technology in Japan, http://www.mext.go.jp/ (accessed 05.04.2012).