Detection of radioactive materials from the Fukushima Daiichi Nuclear Power Plant accident at Shizuoka-city

Makoto Yanaga and Yasuhisa Oya

Radioscience Research Laboratory, Faculty of Science, Shizuoka University, 836 Ohya, Suruga-ku, Shizuoka 422-8529, Japan.

Received May 8, 2012; accepted Feb. 22, 2013

The γ-rays of the atmospheric particles collected in Shizuoka-city from March 15 to March 27, 2011 were measured. Radioactive nuclides due to the Fukushima Daiichi Nuclear Power Plant accident were found in the samples. Day to day variation of concentration of $^{131}$I and $^{137}$Cs etc. indicated two times arrival of radioactive plume from Fukushima to Shizuoka-city and suggested that the surface of Shizuoka-city had been slightly contaminated with radioactive cesium on mainly March 15.

Key words: nuclear power plant accident, atmospheric particles, $^{131}$I, $^{137}$Cs, Shizuoka-city

Introduction

An earthquake of magnitude-9.0 and subsequent huge tsunami hit the Pacific Ocean coast of northeastern Japan on 11 March, 2011. The Fukushima Daiichi Nuclear Power Plant of the Tokyo Electric Power Company located in Fukushima Prefecture was also struck by the tsunami. Although the reactors shut down automatically, the disasters knocked out the cooling systems of the nuclear power plant. Then, a meltdown of the nuclear fuel rods and several times explosions occurred. The accident resulted in a release of radioactive materials, such as $^{131}$I, $^{134}$Cs and $^{137}$Cs etc., into the environments, and formation of high dose rate zones around the Plant and to the northwest direction$^1$.

In order to search for widespread distribution of the radioactive materials, we collected air samples at Shizuoka-city, Japan, about 370 km southwest of the Fukushima Daiichi Nuclear Power Plant, from 15 March, and examined (Fig. 1). On June 2011, Shizuoka Prefecture said that radioactive cesium levels exceeded the safety limit in tea leaves processed at several factories in Warashina area of Shizuoka-city. The tea products had 581 to 654 Bq kg$^{-1}$ of the radioactive cesium, against the Japanese government’s threshold value, 500 Bq kg$^{-1}$, for vegetables$^2$. Therefore, we also discussed on when Shizuoka-city had been contaminated with radioactive cesium without consideration about a question; Was it right to have applied the regulation level for leaf vegetables to processed tea? Tea producers in Shizuoka suffered financial damage not only by not having been able to ship tea but also by the rumor. Many people stopped buying tea leaves processed in Shizuoka because they thought that Shizuoka products were not good for health, regardless of actual safety.

Experimental

Atmospheric particle samples were collected from March 15 to March 27, 2011, on glass-cellulose fiber filters (HE-40T; 48 mm diameter, 0.41 mm thickness, ADVANTEC Co., Ltd.) with an air sampler (SP-30, M&F Enterprise) placed in front of the building of Radioscience Facility at Faculty of Science, Shizuoka University, Shizuoka-city, Japan (Fig. 2). The air sampler used here is usually used for working environment measurements in our radiation controlled area. In the working environment measurements, this sampling device is set at sampling time of 30 minutes beforehand and used for collection of airborne radioactive particles in the laboratories. Therefore, in the present work, the sampling time was set for 30 min at a flow rate of 30 L min$^{-1}$.

The filters were folded in four and wrapped with polyethylene film. The γ-rays of radioactive nuclides in them on 5 mm thick acrylic plate were measured with a coaxial ORTEC GMX series high purity Ge semiconductor detector. The detector was shielded by 100 mm thick lead bricks, 5 mm thick copper plates...
Fig. 1 Locations of sampling and the Fukushima Daiichi Nuclear Power Plant.

Fig. 2 Locations of Shizuoka University and Kita-Ando monitoring site. The topographic map used is publishing by Geospatial Information Authority of Japan.
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and 5 mm acrylic plates. At first, the sample collected on March 15 was placed axially centered 55 mm above the detector Be window and the radioactivity in it was measured. Then, the sample was also measured at 10 mm distance for determination of low counting rate nuclide, $^{131}$I. An efficiency calibration curve at 55 mm from detector surface was determined by measuring commercially obtained calibration sources. The radionuclides employed for the calibration were $^{54}$Mn, $^{57}$Co, $^{60}$Co, $^{109}$Cd, $^{137}$Cs and $^{133}$Ba. An efficiency curve at 10 mm distance was obtained by parallel translation of the efficiency curve at 55 mm using the determined value of $^{131}$I as the secondary standard nuclide. All the samples except for one collected on March 15 were measured at 10 mm above the detector surface. The spectra were stored on a 4096 multi-channel analyzer for 6 - 24 h and were analyzed with the computer software Gamma Studio (SEIKO EG&G Ltd.).

Results and Discussion

Figure 3(a) shows the $\gamma$-ray spectrum of the atmospheric particles collected on March 15. The dominant peaks of the spectrum are those due to $^{132}$Te, $^{131}$I, $^{134}$Cs, and $^{137}$Cs. These nuclides are highly volatility fission products and rather short-lived products, such as $^{132}$Te ($T_{1/2} = 3.20$ d) and $^{131}$I ($T_{1/2} = 8.02$ d), are included in the sample. On the other hand, as shown in Fig. 3(b), no peaks due to these nuclides are found in the spectrum of the sample collected on March 18, three days after the explosion at No. 2 reactor. The spectrum was similar to background spectrum of the detector. These indicate that the nuclides found in the atmospheric particles collected in Shizuoka-city were a part of released radioactive nuclides from the Fukushima Daiichi Nuclear Power Plant. In Fig. 3(a), peaks of $^{131}$I ($T_{1/2} = 2.30$ h), which is daughter nuclide
of $^{135}$Te, and $^{140}$La ($T_{1/2} = 1.68$ d) are also seen in the spectrum whereas $^{146}$Ba ($T_{1/2} = 12.8$ d), the parent nuclide of $^{148}$La, is not recognized. Concentrations of radioactive materials at the end of sampling corrected for decay are summarized in Table 1. In Table 2, concentrations of $^{131}$I, $^{135}$Te, $^{134}$Cs and $^{137}$Cs sampled on March 15 are compared with those of KEK (High Energy Accelerator Research Organization) and RIKEN$^{(1)}$. In the present work, glass-cellulose fiber filters were used in the sampling with no activated charcoal filters and only particles were collected whereas large part of released $^{131}$I existed in gaseous form$^{9}$. The data used in Table 2 for radioactivity concentrations of $^{131}$I are those for particles collected on filters at KEK and RIKEN. KEK in Tsukuba-city, Ibaraki Prefecture, is located 165 km south of the Fukushima Daiichi Nuclear Power Plant and RIKEN in Wako-city, Saitama Prefecture, is about 200 km southwest of the Plant. When a ratio of $^{131}$I for $^{137}$Cs at Shizuoka-city is compared with those of KEK and RIKEN, the ratio of $^{131}$I in Shizuoka-city is much higher whereas the ratio of $^{135}$Te for $^{137}$Cs is approximately constant. According to the observation data of the Japan Meteorological Agency, there was the time when it became cloudy, but the rain was not observed in the daytime from the early morning of March 15 of each Kanto prefecture including Ibaraki and Saitama.$^{6}$ It is hard to think that aerosol-bound radiocesium was removed from the atmosphere by rain on March 15. The data of the Japan Meteorological Agency also indicates that the direction of the wind in each Kanto prefecture from early morning to the daytime of the day blew from Kanto to carry the radioactive plume to Shizuoka. Therefore, the ratio of $^{131}$I having been high in Shizuoka-city indicates that the particle size of radioactive tellurium and cesium is much larger than that of iodine and some quantity of $^{135}$Te, $^{134}$Cs and $^{137}$Cs might fall out before the radioactive plume reached Shizuoka, in the route of detouring around the south side of Mt. Fuji$^{7}$. In fact, the particle size distribution of atmospheric particles of radio nuclides collected in Tsukuba-city showed that much particle size of cesium (both of $^{134}$Cs and $^{137}$Cs) was 2 or 3 µm and most of particles of $^{134}$Cs in solid form were less than 1 µm although large part of $^{131}$I existed in gaseous form$^{9}$. From March 16, only $^{131}$I was detected with lower counting rates due to rather low sampling volume (900 L). Changes in the radiation dose rate measured at Kita-Ando site (see Fig. 2) and meteorological data, precipitation, in Shizuoka-city are shown in Fig. 4.$^{6,8}$ A distinct peak of dose rate on March 15

| Start and end of air sampling | Sampling volume | $^{131}$I (Bq m$^{-3}$) | $^{134}$Te (Bq m$^{-3}$) | $^{134}$Cs (Bq m$^{-3}$) | $^{137}$Cs (Bq m$^{-3}$) | $^{131}$I/$^{137}$Cs | $^{134}$Te/$^{137}$Cs | $^{132}$Cs/$^{137}$Cs |
|-----------------------------|----------------|-------------------------|-------------------------|-------------------------|-------------------------|-----------------|-----------------|-----------------|
| 2011/3/15 16:54 - 17:24     | 0.9 m$^3$      | 26.56±0.43              | 2.19±0.14               | 2.18±0.14               | 17.6±0.6                | 7.4±2.4         |                  |                  |
| 2011/3/16 10:45 - 11:15     | 0.9 m$^3$      | 0.27±0.04               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/17 11:10 - 11:40     | 0.9 m$^3$      | 0.21±0.05               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/18 11:50 - 12:20     | 0.9 m$^3$      | ND                      | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/19 13:30 - 14:00     | 0.9 m$^3$      | ND                      | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/20 12:04 - 12:34     | 0.9 m$^3$      | 0.14±0.04               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/21 7:40 - 8:10       | 0.9 m$^3$      | 0.20±0.05               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/22 11:05 - 11:35     | 0.9 m$^3$      | 0.74±0.06               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/23 10:45 - 11:15     | 0.9 m$^3$      | 0.64±0.04               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/24 11:40 - 12:10     | 0.9 m$^3$      | 0.30±0.05               | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/25               |              |                        |                        |                        |                        |                  |                  |                  |
| 2011/3/26 15:49 - 16:19     | 0.9 m$^3$      | ND                      | ND                      | ND                      | ND                      | ND              |                  |                  |
| 2011/3/27 14:56 - 15:26     | 0.9 m$^3$      | 0.15±0.05               | ND                      | ND                      | ND                      | ND              |                  |                  |

| Detection limit (Bq m$^{-3}$): | 0.08 | 0.04 | 0.04 | 0.2 | 1.0 |

| KEK | $^{134}$Te | $^{134}$Cs | $^{137}$Cs | $^{131}$I/$^{137}$Cs | $^{134}$Te/$^{137}$Cs | $^{132}$Cs/$^{137}$Cs |
|-----|------------|------------|------------|----------------------|----------------------|----------------------|
| Shizuoka Univ. | 26.6 | 17.6 | 2.19 | 2.18 | 12.2 | 8.1 | 1.0 |

| RIKEN | $^{134}$Te | $^{134}$Cs | $^{137}$Cs | $^{134}$I/$^{137}$Cs | $^{134}$Te/$^{137}$Cs | $^{132}$Cs/$^{137}$Cs |
|-------|------------|------------|------------|----------------------|----------------------|----------------------|
|       | 36 | 61 | 7.6 | 9.5 | 3.8 | 6.4 | 0.8 |

Table 2 Concentrations (Bq m$^{-3}$) of $^{131}$I, $^{135}$Te, $^{134}$Cs, and $^{137}$Cs, and their ratios to $^{137}$Cs

Table 1 Radioactivity concentrations of $^{131}$I, $^{134}$Cs, $^{137}$Cs, $^{132}$Te, and $^{140}$La, detected at Shizuoka-city, Japan (Bq m$^{-3}$)
and that from March 21 to March 22 are shown, indicating two times arrival of the dispersed radioactive plume from Fukushima to Shizuoka-city. The fact that there was two time transportation of contaminated air masses to Shizuoka-city in this period was reproduced by numerical simulations\(^7\),\(^9\),\(^10\). Morino et al. and Yasunari et al. also tried reconstruction of atmospheric behavior and deposition of \(^{137}\)Cs using numerical simulations\(^11\),\(^12\). However, there are some discrepancies between the observed and simulated data of deposition because of uncertainties in the treatment of emission, and transport and deposition due to complexities of regional wind and so on. It is impossible to fully reproduce and estimate the distribution of \(^{137}\)Cs deposition across Japan from a limited numbers of observed data.
$^{137}$Cs in the particulate phase can be removed from the atmosphere and brought to the surface by dry or wet deposition. According to the meteorological data, precipitation was observed from March 21 to March 23 in southern Tōhoku and Kanto area\(^6\). Therefore, the majority of aerosol-bound $^{137}$Cs with diameters of several micrometers would be brought to the surface by wet deposition in these areas. In fact, high deposition rates were observed in Yamagata, Ibaraki, Saitama, Chiba, Tokyo, and Kanagawa\(^2\). Then, the second peak of dose rate in Fig. 4 would due to $^{131}$I existed in gaseous form or bound to fine particle. Results of another study on distribution of $^{134}$Cs and $^{137}$Cs in Shizuoka-city also suggest that particles with radioactive cesium released from the Fukushima Daiichi Nuclear Power Plant were arrived at Shizuoka-city on mainly March 15, 2011\(^1\). Surface of Shizuoka-city would be slightly contaminated with radioactive cesium on mainly March 15, 2011.

**Conclusion**

In the present work, atmospheric particle samples were collected from March 15 to March 27, 2011, after the Fukushima Daiichi Nuclear Power Plant accident occurred, and radioactivity in the samples was determined. Radioactive nuclides from Fukushima were found in the samples. Day to day variation of concentration of $^{132}$Te, $^{131}$I, $^{134}$Cs, and $^{137}$Cs indicated two times arrival of radioactive plume from Fukushima and suggested that the majority of radioactive cesium on the surface of Shizuoka-city had been deposited on mainly March 15.

**Acknowledgement**

We appreciate the financial support from Shizuoka University, Japan. We are grateful to Mr. Toshiyoshi Miyazawa (Faculty of Science, Shizuoka University) for his help in the sampling of atmospheric particles.

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