Measurement of radioactivity in airborne dust and estimation of public dose in Tokyo after the Fukushima Daiichi Nuclear Power Plant accident

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We report on the results of emergency monitoring of environmental radiation in Tokyo after the severe accident at the Fukushima Daiichi Nuclear Power Plant, triggered by the Great East Japan Earthquake and the resulting tsunami on the 11th March 2011. We collected airborne dust samples at the Setagaya and Koto districts of Tokyo. The concentrations of radionuclides were measured by a Ge semiconductor detector. The highest concentrations of major fission products were detected at 10:00–11:00 a.m. on the 15th March 2011. The detected concentrations were: 2.4×10^{2} Bq/m^3 of ^{131}I, 6.4×10^{1} Bq/m^3 of ^{134}Cs, and 6.0×10^{1} Bq/m^3 of ^{137}Cs. The concentration of ^{131}I fell to undetectable levels by June 2011. In contrast, ^{134}Cs and ^{137}Cs were detected intermittently for one year. Based on the results on the concentrations of detected nuclides, the public internal doses due to inhalation were estimated on the assumption that those in the affected area stayed outdoors continuously for the period from the 13th March 2011 to the 12th March 2012 in the Tokyo metropolitan area. The committed effective dose due to inhalation for adults was estimated to be 25 μSv.

Key Words: *Fukushima Daiichi NPP, radiation monitoring, committed effective dose*

1. Introduction

The Great East Japan Earthquake and the resulting tsunami on the 11th March 2011 triggered a severe accident at the Fukushima Daiichi Nuclear Power Plant (NPP). The Tokyo Metropolitan Industrial Technology Research Institute (TIRI) has been in charge of environmental radiation monitoring since 1975. This monitoring is based on the 'Regional Disaster Prevention Programme' formulated by the Tokyo Metropolitan Government. On the 13th March, two days after the Fukushima Daiichi NPP disaster, emergency monitoring of the environmental radiation to detect gamma-emitting nuclides in airborne dust was initiated.

This paper describes the results of the monitoring of gamma-emitting nuclides concentrations in airborne dust for the period from the 13th March 2011 to the 12th March 2012 in the Tokyo metropolitan area. Based on these results, we estimated the public internal doses due to inhalation. The radionuclide concentrations measured in the Tokyo metropolitan area were reported on the publicly accessible website of the Bureau of Industrial and Labor Affairs, Tokyo Metropolitan Government[1)].

2. Material and methods

(2.1) Sampling of airborne dust

Our sampling conditions were based upon the guidelines presented in reference [2]. Airborne dust samples were collected at the TIRI facility in the Setagaya (13th March

2011–12th October 2011) and Koto (13th October 2011–12th March 2012) districts of Tokyo, which are located about 220 km and 230 km southwest of the Fukushima Daiichi NPP, respectively. Airborne dust samples were collected with a glass-fiber filter (GB-100R, Advantec, Dublin, CA) using the two high-volume air sampler models: TFIA (Staplex Co., Brooklyn, NY) from the 13th March 2011 to the 10th April 2011; and HV-500R (Sibata Scientific Technology Ltd., Saitama, Japan) from the 14th April 2011 to the 12th March 2012. The air samplers were set at a height of about 1 m above ground level. The air flow rate was set as 0.6 m^3/min. The interval between filter replacements varied between 1 h and 24 h according to the air dose rate.

(2.2) Measurement of concentrations of radionuclides

The radioactivity of the collected samples was measured by a germanium (Ge) semiconductor detector which used the GR3019 (Canberra Industries, Meriden, CT) for the period of the 13th March 2011 to the 10th April 2011, and the GMX Series 25 (Ortec, Oak Ridge, TN) for the period of the 11th April 2011 to the 12th March 2012. The measurement time varied between 1,000 s and 20,000 s according to the air dose rate. The counting efficiency of the Ge semiconductor detector was calibrated using a standard radiation source supplied by the Japanese Radioisotope Association. The coincidence summing was corrected, measurement of each sample was carried out immediately after the sampling, and the decay corrections of radionuclides were not considered.

To examine the correlation between the concentrations of radiocaesium (Bq/m^3) and airborne dust (mg/m^3), we weighed the airborne dust samples adsorbed on the filter (April 19th–October 20th). The airborne dust samples' weights were measured under constant conditions where the temperature and the relative humidity were regulated at 20°C and 50 %, respectively.

3. Results and Discussion

(3.1) Concentrations of gamma-emitting nuclides in airborne dust samples

The changes in the concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs are shown in Figs. 1 and 2. The peak that first detected fission products was observed in the sample collected at 0:00–7:12 a.m. on the 15th March 2011 at Setagaya. The

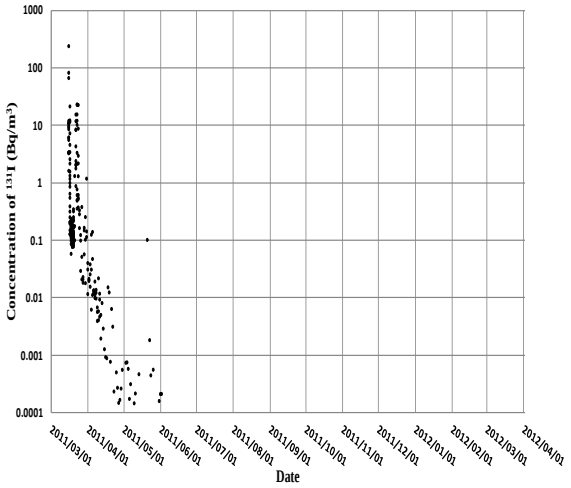

Fig. 1. Changes in the concentration (Bq/m^3) of ^{131}I in airborne dust samples for the period from the 13th March 2011 to the 12th March 2012 in Tokyo.

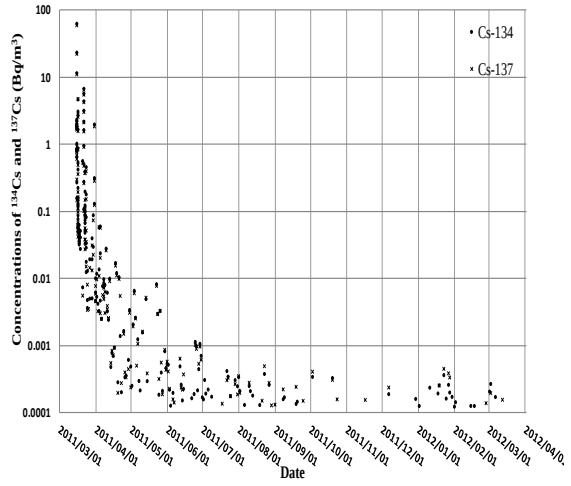

Fig. 2. Changes in the concentrations (Bq/m^3) of ^{134}Cs and ^{137}Cs in airborne dust samples for the period from the 13th March 2011 to the 12th March 2012 in Tokyo.

concentrations were: 1.1×10^1 Bq/m^3 of ^{131}I, 1.9 Bq/m^3 of ^{134}Cs, and 1.8 Bq/m^3 of ^{137}Cs. After the initial detection of fission products, we started to collect samples at intervals of 1 h. The concentrations had decreased in the sample taken at about 8:00 a.m., and then increased again at about 9:00 a.m. A second peak demonstrating the highest concentrations of radionuclides was detected at 10:00–11:00 a.m. of the same day (15th March). The concentrations were: 2.4×10^2 Bq/m^3 of ^{131}I, 6.4×10^1 Bq/m^3 of ^{134}Cs, and 6.0×10^1 Bq/m^3 of ^{137}Cs. The gamma-ray spectrum is shown in Fig. 3. Twelve gamma-emitting nuclides can be identified in the spectrum. The detected

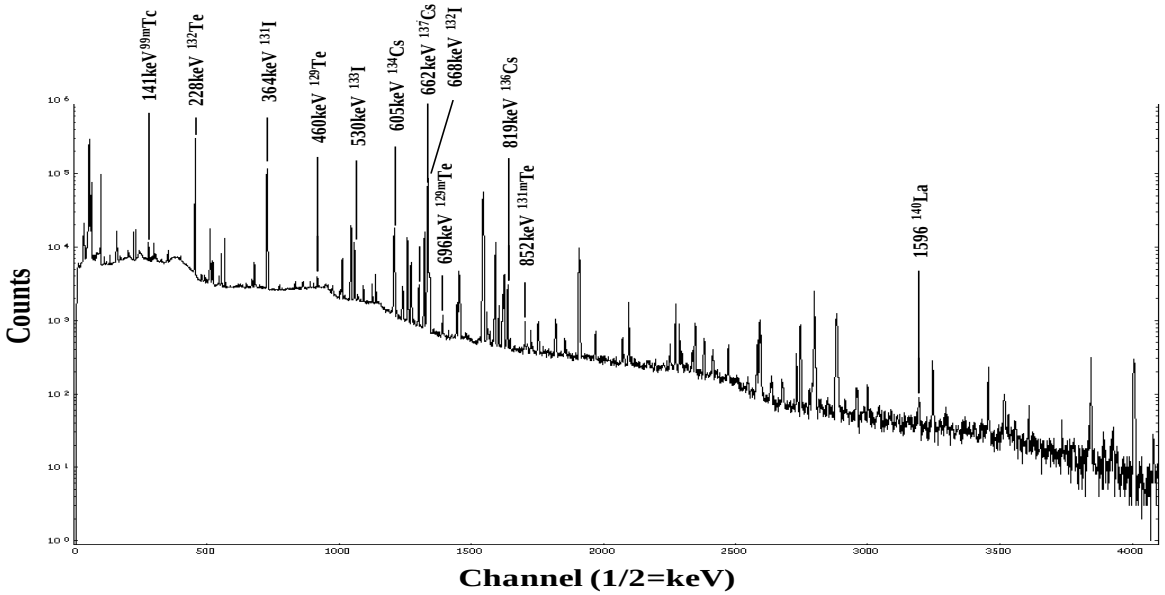

Fig. 3. The gamma-ray spectrum of the samples collected at 10:00–11:00 a.m. on the 15th March 2011. The peaks that were used to calculate the radionuclide concentrations are indicated.

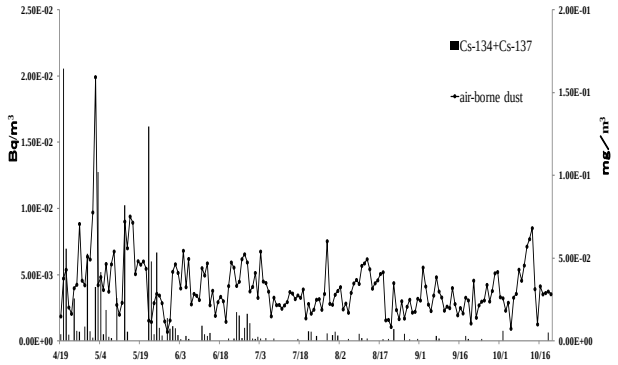

Fig. 4. The change in the concentrations of radiocesium (Bq/m^3) and airborne dust samples (mg/m^3). The concentration of radiocaesium shown here is the sum of ^{134}Cs and ^{137}Cs.

radionuclides (and their half-lives) were as follows: ^{99m}Tc (6.0 h), ^{129m}Te (34 d), ^{129}Te (70 m), ^{131m}Te (30 h), ^{132}Te (3.2 d), ^{131}I (8.0 d), ^{132}I (2.3 h), ^{133}I (21 h), ^{134}Cs (2.1 y), ^{136}Cs (13 d), ^{137}Cs (30 y) and ^{140}La (1.7 d). A third peak was observed in samples taken at 4:00–5:00 a.m. of the following day (16th March 2011). The concentrations were: 2.2×10^1 Bq/m^3 of ^{131}I, 4.7 Bq/m^3 of ^{134}Cs, and 4.8 Bq/m^3 of ^{137}Cs. The concentrations then decreased and leveled off to almost undetectable levels, increasing again on the 21st–23rd March, presumably due to rainfall. The concentration of ^{131}I had fallen to an undetectable level in June 2011, whereas the concentrations of ^{134}Cs and ^{137}Cs were detected intermittently for one year.

The changes in the concentrations of radiocaesium (Bq/m^3) and airborne dust samples (mg/m^3) with time, are shown in Fig. 4. As seen in the figure, there was no correlation between the concentration of radiocaesium and the concentration of airborne dust. This lack of a correlation might indicate that the concentrations in airborne dust samples do not contribute to the concentrations of radionuclides.

(3.2). Committed effective dose estimation due to inhalation

The committed effective dose due to inhalation was estimated from our results. The dose was calculated by the following equation:

$$H = \left(\sum_i Ki \cdot Xi \right) \cdot M \cdot T$$

where H is the committed effective dose (μSv); Ki is the effective dose coefficient; Xi is the air concentration of the radionuclide; M is the inhalation rate; and T is the period of exposure. The dose estimation is referred to in the guideline given in reference [3]. Ki and M were cited as they appeared in the International Commission on Radiological Protection (ICRP) Publications 71[4] and 72[5], where it was assumed that those in the affected area

Table. 1. The committed effective dose of adults due to inhalation for the period of the 13th March 2011 to the 12th March 2012. "Other nuclides" includes ^{99}Mo-^{99m}Tc, ^{129m}Te-^{129}Te, ^{131m}Te, ^{132}Te-^{132}I, ^{133}I, ^{136}Cs, and ^{140}Ba-^{140}La.

Nuclides	Committed effective dose (μSv)			
	3/13, 2011–3/31	4/1–5/31	6/1–3/12,2012	Total
^{137}Cs	8.4	0.16	0.01	8.6
^{134}Cs	4.5	0.08	0.01	4.6
^{131}I	7.6	0.05	> 0.00	7.7
Other nuclides	3.7	> 0.00	> 0.00	3.7
Total	24.2	0.29	0.02	25

stayed outdoors continuously.

The results of the committed effective dose calculation are shown in Table 1. ^{131}I, ^{134}Cs, and ^{137}Cs were the major radionuclides in the calculated effective dose. Most of the total dose was given in March after which the dose decreased to nearly background levels. The committed effective dose of adults was estimated to be 25 μSv. The actual dose may be rather higher than that estimated here if internal doses are taken into consideration, for example, by measuring the ratio of gaseous iodine to total iodine, as reported by Amano et al.[6] and Takeyasu et al.[7].

4. Conclusions

In response to the accident at the Fukushima Daiichi NPP, we performed emergency environmental radiation monitoring at TIRI. The concentrations of radionuclides were measured during the period from the 13th March 2011 to the 12th March 2012 in Tokyo. The highest concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were 2.4×10^{2} Bq/m^3, 6.4×10^{1} Bq/m^3, and 6.0×10^{1} Bq/m^3, respectively. Based on these results, we estimated that the committed effective dose due to inhalation for adults in the Tokyo metropolitan area was 25 μSv.

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