Radioactivity of the aerosol collected in Nagasaki City due to the Fukushima Daiichi Nuclear Power Plant Accident

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Abstract

Radioactivity of ^{134}Cs and ^{137}Cs was detected in the aerosol collected in Nagasaki prefectural forest park "Nagasaki Kenmin No Mori" about 20 km north-west from central Nagasaki City from Mar. 23 to Jul. 27, 2011. The highest concentrations of the nuclides were detected in the sample collected from Apr. 6 to Apr. 13 and ^{110m}Ag was also detected in the sample. The wind of Apr. 6 in the park was found to come via Fukushima with back-trajectory analysis in the web-site of National Oceanic Atmospheric Administration (NOAA), United States Department of Commerce. The concentrations in the air of ^{134}Cs, ^{137}Cs and ^{110m}Ag evaluated were as small as 0.47, 0.52 and 0.0054 mBq/m^3 respectively. However, the concentrations of them in the collected aerosol were as large as 11.3, 12.4 and 0.12 kBq/kg, and equivalent to the level of surface soil of 5 cm in Warabidaira Iitate Fukushima, highly contaminated area. It indicates that air filters in air-conditioning facilities should be handled carefully also at Nagasaki about 1,000 km apart from Fukushima Daiichi Nuclear Power Plant. In addition, the concentration of natural radioactivity Pb-210 was found as large as 19.9 kBq/kg. Therefore, it was ascertained that the risk of air filters was already existed before the accident and the radioactivity arisen from the accident increased the risk.

INTRODUCTION

The nuclear accident at the Fukushima Daiichi Nuclear Power Plant occurred on 11 March 2011.

Many types of radioactive nuclides, ^{129m}Te, ^{129}Te, ^{131}I, ^{132}Te, ^{132}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{99m}Tc, ^{95}Nb and ^{110m}Ag were detected from soil samples collected near the nuclear power plant from 15 March.[1)] Detection of ^{99m}Tc, ^{95}Nb and ^{110m}Ag were not reported in the paper in the sake of some measurement difficulty. Now the detection has been confirmed.

We are continuing to measure various new samples sent from Fukushima with the request of the measurement of the people and for the purpose of study. However, detection of the nuclides other than ^{134}Cs, ^{137}Cs and ^{110m}Ag became difficult now because of these short half-lives.

Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japanese government presented the distribution maps of radioactivity concentration of ^{134}Cs, ^{137}Cs, ^{129m}Te and ^{110m}Ag around the nuclear power plant[2, 3, 4)]. These maps show the distribution of the nuclides on land in the range of 100 km from the power plant. MEXT is also presented distribution map in the seawater around the power plant[5)]. They indicated that ^{131}I, ^{134}Cs and ^{137}Cs were not detected in all the seawater samples except at the point just in front of the power plant. However, we detected ^{134}Cs and ^{137}Cs from seawater of Hisanohama, about 30 km south of the power plant. The radionuclides were also detected in the sediment of the seabed. Higher concentrations than the seawater of ^{134}Cs, ^{137}Cs and ^{110m}Ag were found in abalones and sea urchins.

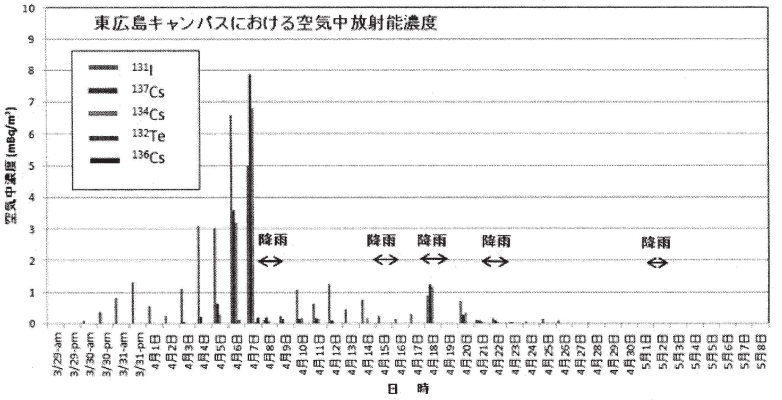

Fig. 1. Radioactivity of aerosol collected in Higashihiroshima Campus, Hiroshima University by Shizuma[6]. The figure was reprinted from the reference. Horizontal axis is sampling time written in Japanese. For example, "4 月 7 日" means April 7. Vertical axis is radioactivity of the aerosol per air volume. Arrows indicate rainfall. Largest radioactivity was detected on April 7.

^{110m}Ag concentration of the abalones and sea urchins was higher than the seabed sediments near the sampling points . These were reported in a television program of Japan Broadcasting Corporation (NHK) at November 27, 2011.

Shizuma[6] reported that ^{131}I, ^{137}Cs, ^{134}Cs ^{132}Te and ^{136}Cs were detected in the aerosol collected in Higashihiroshima Campus, Hiroshima University. The concentration was largest on April 7, 2011 (Fig. 1).

We now present that the radioactive materials due to the nuclear power plant accident have been spread out also to Nagasaki city about 1,000 km from the power plant.

MATERIALS AND METHODS

Sampling of aerosol

A high volume air sampler (Shibata AH600-F) has been installed at Nagasaki prefectural forest park "Nagasaki Kenmin No Mori" (Fig. 1) about 20 km north-west from central Nagasaki City intended for study of transboundary air pollution across East China Sea.

Aerosol was collected with quartz fiber filters (Advantec QR-100). The mass of collected aerosol was estimated subtracting mass of the filter before collecting aerosol from the mass after collecting. The mass of the filter was

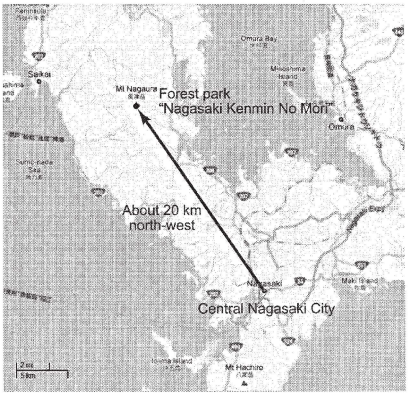

Fig. 2. Nagasaki prefectural forest park "Nagasaki Kenmin No Mori

weighed after keeping 30～40%RH of humidity in a desiccator cabinet (Sanplatec Type A-3) because the mass depend on the humidity.

Measurement of Radioactivity in the aerosol

The filter was pressed into a disk in a die with a press machine. Radioactivity was measured with a germanium detector (Ortec GMX30). The detection efficiency was determined using mixed large volume calibration standards (Isotope products laboratories EG-ML) containing radionuclides of ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{88}Y and ^{60}Co, the uncertainty of the activities was certified less than 5%, samples of natural uranium rich grinded rock and ^{210}Pb rich surface soil samples collected near Nagasaki University to determine detection efficiency of ^{210}Pb.

Air Trajectory Analysis

Air trajectory analysis was performed in the web-site of National Oceanic Atmospheric Administration (NOAA), United States Department of Commerce[7]. In the web page, link of "Run HYSPLIT Trajectory Model" was selected in the category of HYSPLIT-WEB (Internet-based). In the web page opened, link of "Compute archive trajectories" was selected. In the web page opened, "Next" button was clicked with the default settings (Number of Trajectory Starting Locations: 1, Type of Trajectory: Normal) in the web page. "GDAS" was selected as meteorological data. Maps of air trajectory were obtained by setting appropriate values in the subsequent web pages.

RESULT

Sampling and Measurement of Radioactivity

Table 1 shows sampling period, mass of collected aerosol, air volume introduced in the air sampler, date and measurement time. Table 2 shows radioactivity of the collected aerosol per the introduced air volume. The values of ^{134}Cs, ^{137}Cs, ^{110m}Ag and ^{210}Pb are plotted in Fig. 3. The

Table 1. Mass of collected aerosol, air volume, date and measurement time

Sampling period	Mass of collected aerosol (mg)	Air volume (m^3)	Date measurement was started	Measurement time (h)
02 Mar - 09 Mar	116	7,035	2-May	48
09 Mar - 16 Mar	116	7,035	21-Apr	48
16 Mar - 23 Mar	86.3	6,922	19-Apr	48
23 Mar - 30 Mar	304	7,032	18-Apr	24
30 Mar - 06 Apr	302	7,119	17-Apr	24
06 Apr - 13 Apr	296	6,974	7-Jul	165
13 Apr - 20 Apr	220	7,185	14-Jul	49
20 Apr - 27 Apr	142	6,917	16-Jul	72
20 Jul - 27 Jul	115	7,186	28-Jul	24
27 Jul - 03 Aug	77.4	6,852	8-Aug	46
03 Aug - 10 Aug	66.0	7,070	12-Aug	71
10 Aug - 18 Aug	105	8,023	22-Aug	25

Table 2. Radioactivity in aerosol per air volume. "±" indicate standard error expected from the γ-ray spectrometry.

Sampling period	Concentration ($\times 10^{-6}$ Bq/m^3)					
	^{134}Cs	^{137}Cs	^{110m}Ag	^{210}Pb	^{7}Be	^{40}K
02 Mar - 09 Mar	—	—	—	2349 ± 29	—	—
09 Mar - 16 Mar	1.3 ± 1.2	—	1.8 ± 1.6	1084 ± 22	6733 ± 59	29 ± 15
16 Mar - 23 Mar	—	3.5 ± 1.2	1.7 ± 1.7	750 ± 19	4792 ± 47	47 ± 14
23 Mar - 30 Mar	7.4 ± 1.6	5.9 ± 2.0	—	1184 ± 32	6398 ± 72	59 ± 21
30 Mar - 06 Apr	26.70 ± 0.49	299.2 ± 5.7	—	1040 ± 33	7974 ± 79	40 ± 22
06 Apr - 13 Apr	477.7 ± 2.2	523.6 ± 2.4	5.38 ± 0.99	842 ± 11	4903 ± 45	142.8 ± 7.7
13 Apr - 20 Apr	37.1 ± 1.5	39.6 ± 1.6	—	701 ± 17	3811 ± 64	133 ± 13
20 Apr - 27 Apr	23.2 ± 1.0	29.5 ± 1.1	—	687 ± 12	3358 ± 46	136 ± 11
20 Jul - 27 Jul	5.9 ± 1.6	5.2 ± 1.6	1.9 ± 1.6	322 ± 22	976 ± 59	—
27 Jul - 03 Aug	2.0 ± 1.2	2.1 ± 1.2	—	116 ± 14	1426 ± 22	129 ± 14
03 Aug - 10 Aug	—	—	1.2 ± 1.0	202 ± 12	872 ± 14	134 ± 12
10 Aug - 18 Aug	—	2.5 ± 1.3	—	187 ± 16	1305±26	152 ± 17

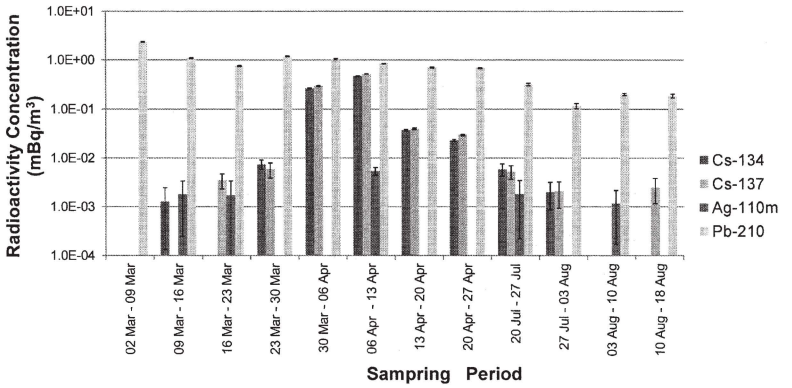

Fig. 3. Radioactivity in aerosol per air volume plotted against sampling period.

sample of the period "06 Apr –13 Apr" showed highest value of ^{134}Cs, ^{137}Cs and ^{110m}Ag. Table 3 shows the radioactivity of the aerosol per mass of the aerosol itself. The values are also plotted in Fig. 4. The values became rather large because the mass of the aerosol is small.

Air Trajectory Analysis

Fig. 5 is the results of backward trajectory analysis from "Nagasaki Kenmin No Mori". The wind from 12:00 UTC 06 to 0:00 UTC 07 was found to come via Fukushima. The wind was found not to come via Fukushima for other time in the sampling period by surveying with the analysis. Fig. 6 is the results for Higashihiroshima Campus , Hiroshima University. The wind from 18:00 UTC 06 to 12:00 UTC 07 was found to come via fukushima. The wind was also found not to come via Fukushima for other time in the sampling period. Fig. 7 is the results of forward trajectory

Table 3. Activity of radionuclides per aerosol mass

Interval of Sampling	Concentration ($\times 10^3$ Bq/kg)					
	^{134}Cs	^{137}Cs	^{110m}Ag	^{210}Pb	^{7}Be	^{40}K
02 Mar - 09 Mar	—	—	—	142.5 ± 1.7	—	—
09 Mar - 16 Mar	0.079 ± 0.071	—	0.110 ± 0.095	65.7 ± 1.3	408.5 ± 3.6	1.74 ± 0.93
16 Mar - 23 Mar	—	0.281 ± 0.095	0.14 ± 0.13	60.2 ± 1.6	384.6 ± 3.8	3.8 ± 1.1
23 Mar - 30 Mar	0.172 ± 0.038	0.136 ± 0.046	—	27.43 ± 0.73	148.2 ± 1.7	1.39 ± 0.48
30 Mar - 06 Apr	6.30 ± 0.12	7.06 ± 0.13	—	24.56 ± 0.73	188.2 ± 1.9	0.95 ± 0.52
06 Apr - 13 Apr	11.273 ± 0.053	12.357 ± 0.058	0.127 ± 0.023	19.87 ± 0.25	115.7 ± 1.1	3.37 ± 0.18
13 Apr - 20 Apr	1.21 ± 0.049	1.29 ± 0.051	—	22.91 ± 0.55	124.5 ± 2.1	4.33 ± 0.42
20 Apr - 27 Apr	1.13 ± 0.051	1.44 ± 0.054	—	33.48 ± 0.58	163.7 ± 2.3	6.62 ± 0.53
20 Jul - 27 Jul	0.37 ± 0.10	0.33 ± 1.6	0.12 ± 0.10	20.2 ± 1.4	61.2 ± 1.7	—
27 Jul - 03 Aug	0.18 ± 0.10	0.19 ± 0.10	—	10.3 ± 1.3	126.2 ± 1.9	11.4 ± 1.3
03 Aug - 10 Aug	—	—	0.13 ± 0.11	21.6 ± 1.2	93.4 ± 1.5	14.4 ± 1.3
10 Aug - 18 Aug	—	0.19 ± 0.10	—	14.3 ± 1.2	99.7 ± 2.0	11.7 ± 1.3

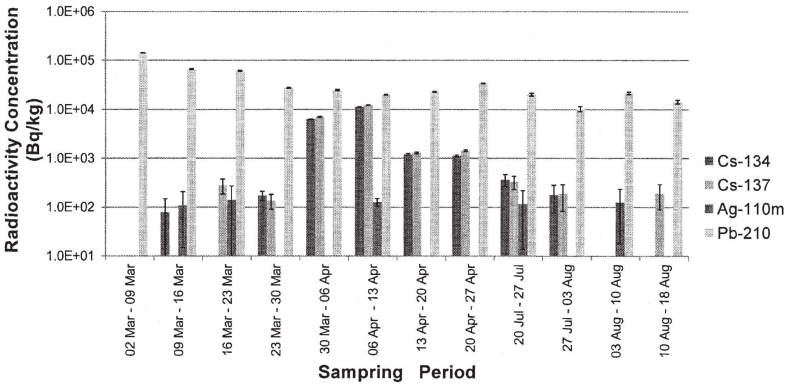

Fig. 4. Radioactivity in aerosol per mass plotted against sampling period.

analysis from Fukushima Daiichi Nuclear Power Plant. The route of air flow was found to move from west to east in the two hours. The wind going through Nagasaki at first changed to the route thorough Hiroshima.

DISCUSSION

It was found that the radioactivity of aerosol became highest when the wind came via Fukushima in both "Nagasaki Kenmin No Mori" and Higashihiroshima. It indicates that the radioactivity originated from Fukushima Daiichi Nuclear Power Plant surely has spread to Nagasaki and Hiroshima.

ICRP Publ. 23 estimates that respiratory volume of reference man per day is 22.21m^3. Dose coefficients of ^{134}Cs, ^{137}Cs and ^{110m}Ag for inhalation intake are 2.0×10^{-8}, 3.9×10^{-8} and 1.2×10^{-8} Sv/Bq according to

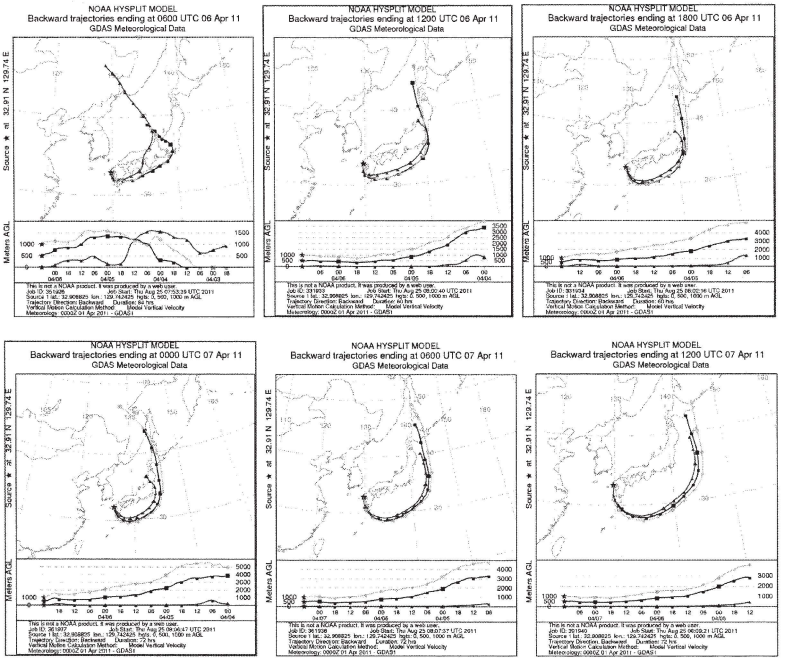

Fig. 5. Backward trajectory analysis of air flow from "Nagasaki Kenmin No Mori" at 6:00, 12:00 and 18:00 UTC 06 and 0:00, 6:00 and 12:00 UTC 07 Apr 2011. The wind from 12:00 06 to 0:00 07 was found to come via Fukushima.

ICRP Publ. 72. Using these values, the committed dose equivalent for a person breathing the air of "Nagasaki Kenmin No Mori" all in the sampling intervals was calculated as 0.008 μSv. Therefore, the radiation dose may be negligible compared with the dose of natural radiation even postulating large uncertainty for these estimations. However, the mass concentration of ^{134}Cs and ^{137}Cs of aerosol itself was awfully high and the value was equivalent to the level of surface soil of 5 cm in Warabidaira Iitate Fukushima, highly contaminated area[1]. It indicates that cautions should be exercised for some cases for example handling of air filters in air-conditioning facilities of some big buildings also at Nagasaki about 1,000 km apart from Fukushima Daiichi Nuclear Power Plant. However, radioactivity of ^{210}Pb was higher than ^{134}Cs and ^{137}Cs. Therefore it is shown that the aerosol was dangerous before the accident with natural radioactivities and ^{134}Cs and ^{137}Cs only increases the dangerousness.

ACKNOWLEDGEMENT

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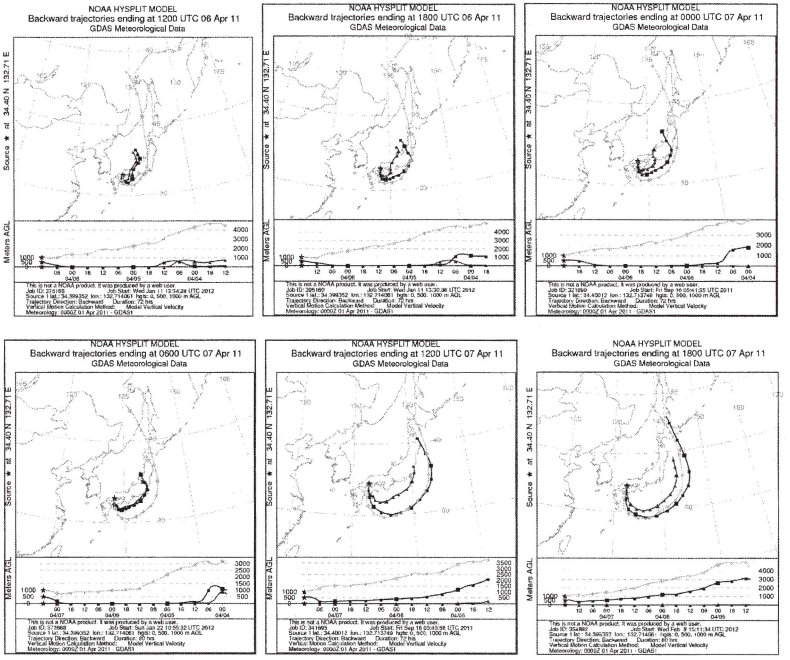

Fig. 6. Backward trajectory analysis of air flow from Higashihiroshima Campus, Hiroshima University at 12:00, 18:00 UTC 06 and 0:00, 6:00, 12:00, 18:00 UTC 07 Apr 2011. The wind from 18:00 06 to 12:00 07 was found to come via fukushima.

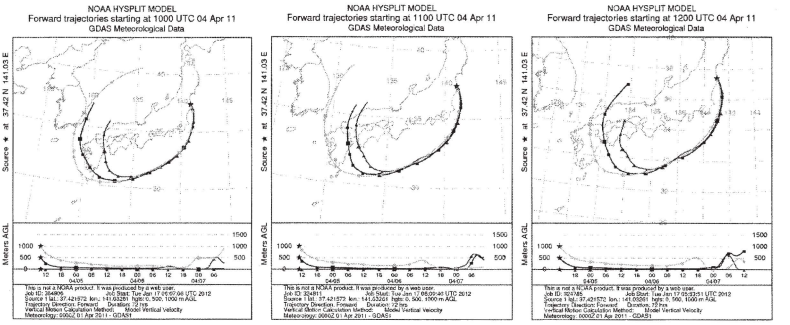

Fig. 7. Forward trajectory analysis of air flow from Fukushima Daiichi Nuclear Power Plant, at 10:00, 11:00 and 12:00 UTC 04 Apr 2011. The route of air flow was found to move from west to east in the two hours.

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