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Measurement of ^{134,137}Cs Concentrations in the Air of Fukushima-City

By

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Abstract: Concentrations of radioactive Cs isotopes in Fukushima-City were measured by collecting the air dust on the air filter for automobile on Oct. 21 and 22, 2011. Gamma-rays from the filter paper were counted by a NaI scintillation counter and a Ge detector. Dividing the activities by the volume of air passed through the filter, the concentrations of ^{134}Cs and ^{137}Cs were estimated to be 6.13×10^{-4} Bq/m^3 and 8.29×10^{-4} Bq/m^3 respectively, both with 31% statistical error.

Key words: Fukushima Daiichi Nuclear Accident, radioactive cesium, gamma-ray spectrum, atmospheric concentration

1. Introduction

Severe accident in Fukushima Daiichi Nuclear Power Plant after the big earthquake and tsunami on March 11th this year caused a heavy radioactive pollution over a vast area of Japan's soil, including the capital city of Fukushima Prefecture. Radiation level in Fukushima-City is still high, lingering round 1 μ Sv/h. Measurements of radiation levels everywhere and concentrations of radioactive nuclides in everything are essential to protect people from the radiation and radioactivity. Dose rates due to gamma-rays from activities on the ground are widely measured and mapped, as well as the contaminations of the earth. However the data on the pollution of the air in Fukushima Prefecture and near-by areas are rather sparse. The only report we found for Fukushima City is from the University of Fukushima[1]. Another systematic monitoring of nuclides in the air is performed by the CTBT radionuclide observatory station in Takasaki, which is located more than 200km from Fukushima Daiichi, and the data are published regularly on their website[2]. According to these observations, even today, more than seven months after the accident, considerable atmospheric concentrations of Cs isotopes are continuously observed.

Since one of the author K. Toyoshima had an opportunity to visit Fukushima City last October, he tried to collect dust in the air to measure its radioactivity concentration. For this purpose, a simple air dust collector was devised by combining an air filter element for a car and a small fan. This kind of filter has a rather large mesh, but they are easily obtained. In addition a great number of this "collectors" are always working on the roads. So this kind of filter could be a standard device for widely monitoring the radioactivity in the air.

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2. Collection of the air dust

The air filter unit used is "SU-102" of Nitto Kogyo Co. Ltd, a cylindrical one for a small car. The outer diameter is 120mm and the length is 130mm. Small fan, Model "AD0812HS-A70GS" of ADDA CORP was attached to one end and the other end was closed by a wooden board. Both battery and AC adapter were prepared to drive the fan continuously.

Performance detail of this filter is not known, but according to a literature[3] this kind of filters "capture more than 98% of 7 to 9 μ m diameter particles". The power of the fan is 65.59m^3/h or 38.6 CFM.

This device was run continuously throughout the stay of Toyoshima in Fukushima City, starting at 15:56 on Oct. 21, 2011, and terminating at 10:16 next day, carried always by him. Total running time is 18 hours and 30 minutes, which means the air of volume 1.2134×10^3 m^3 in total passed through this filter. Places where this device traveled were: JR Fukushima station, JR Kanayagawa station, Fukushima University, and finally the central area of Fukushima City within 1km from JR station - lyceum, pub, and hotel.

3. Activity measurement of the air filter

The filter paper of the unit was then stripped off and the gamma-rays from it were observed by a NaI scintillation counter and a Ge detector. It was wound

around the side faces of these detectors, which are both cylindrically shaped. The NaI counter is Bicron 2M2/2PR, whose crystal size is 2"ϕ×2". To reduce the gamma-ray background, sample room and the detector itself were enclosed by 11 lead bricks. The Ge detector is the equipment of the Radioisotope Institute for Basic and Clinical Medicine of Kurume University and we were granted to use it.

Gamma-ray spectrum taken by the NaI counter is shown in fig.1, with the background data normalized to the same live-time (active time of the multichannel pulse–height analyzer) as the measurement of the sample. Background spectrum was taken by putting new filter unit (but different lot to the used one). Gamma-ray peaks from ^{134,137}Cs are fitted by a software FitzPeaks (Demo and NaI version) [(4)]. Efficiency calibration was done by putting a calibrated ^{137}Cs source of LMRI (Laboratoire de Métrologie des Rayonnements Ionisants) at several points around the NaI crystal. Considering the geometry of the sample wound around the NaI, the position of 11mm from the cylindrical face and 20.4 mm backward (to the photomultiplier's side) from the front face was chosen as the typical point, and the efficiency there was obtained by averaging the nearest 2 points actually measured.

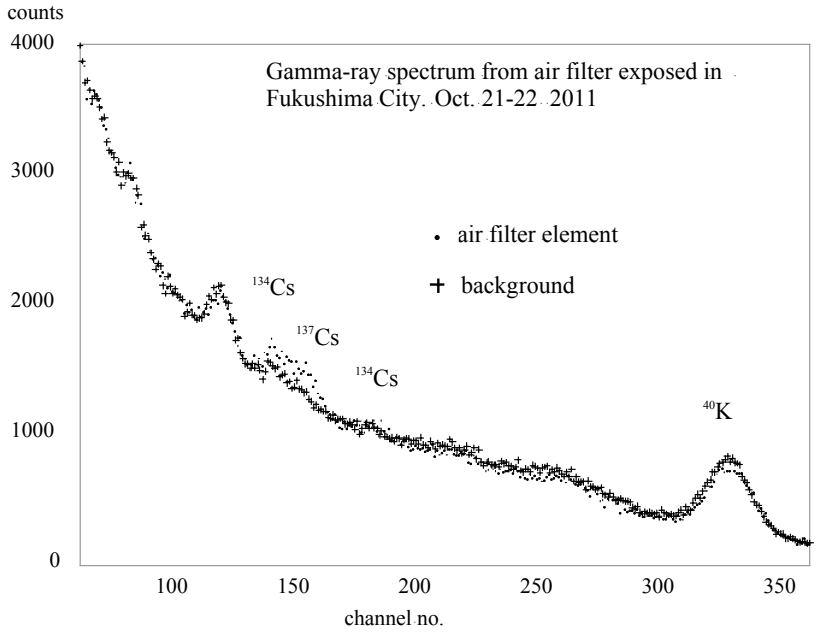

Fig. 1. Gamma-ray spectra taken by NaI counter.

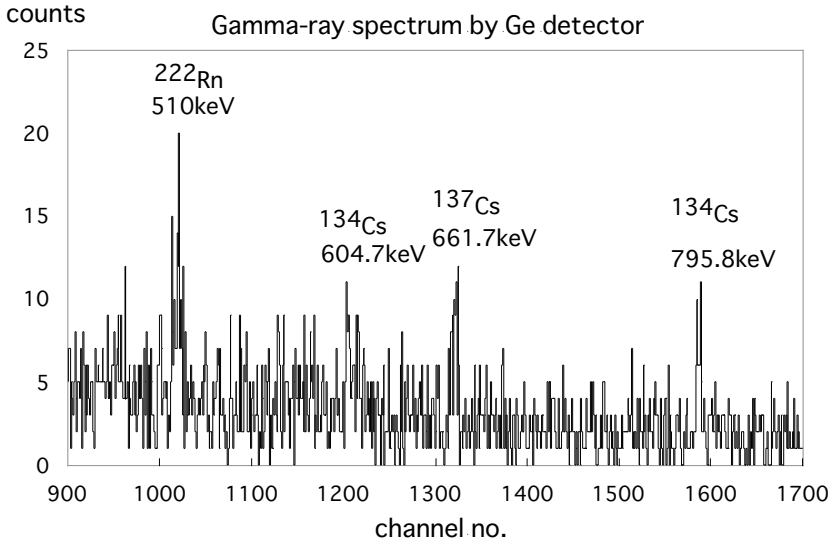

Fig. 2. Gamma-ray spectra taken by the Ge detector.

Measurement and analysis by the Ge detector were performed automatically by the attached computer and software. Three peaks from ^{134}Cs and ^{137}Cs are clearly visible, as shown in fig.2. Since the efficiency of this detector was available only for the front surface of the detector but the sample geometry is complex, absolute activities cannot be known. However, since the relative efficiencies across gamma-ray energies are meaningful, activities of each peaks were evaluated by the efficiencies at the front surface. Results of both detectors are summarized in table 1.

4. Estimation of atmospheric concentration and comparison to other observations

If we divide the activity values in table 1 by the air volume of 1,213.4 m^3 which passed through the filter, we get the concentrations of ^{134}Cs and ^{137}Cs as 6.13×10^{-4} Bq/m^3 and 8.29×10^{-4} Bq/m^3 respectively. They are about ten times greater than the record of Takasaki observatory, which reads 0.73 and 0.98 respectively in the unit of 10^{-4} Bq/m^3, for the intake of Oct. 21 to 22. This difference may reflect the difference in distance from the Fukushima Daiichi between these two observing positions. Considering the roughness of mesh of our filter, these ratios may be greater.

Table 1 Gamma-rays from the filter paper observed by NaI and Ge detectors.

Nuclide	energy (keV)	peak area	error	efficiency (%)	activity (Bq)
NaI detector. live time :167,465s					
^{134}Cs	604.7	2,576	796	2.43	0.744
^{137}Cs	661.65	2,747	838	1.917	1.006
Ge detector. live time : 7,200s					
^{134}Cs	604.7	32.1	13.6	3.27	0.140*
^{134}Cs	795.85	39.6	9.2	2.57	0.251*
^{137}Cs	661.65	57.9	14.2	3.01	0.315*

* Relative values are only meaningful because of the lack of absolute efficiency (see text).

Fukushima University measured radioactive materials in the atmosphere of their own place from May 18 to July 15 with their high-volume air sampler[(1)]. They observed a highest value of 3.29×10^{-5} Bq/l, namely 3.29×10^{-2} Bq/m^3 for total Cs on May 23. A member of this observation group, Professor A. Watanabe gave us their recent data[(5)] observed on Sept. 2, 6 and 10, which show sharp decreases from 6.89 to 1.78 for ^{134}Cs, from 8.01 to 2.11 for ^{137}Cs, all in the unit of 10^{-4} Bq/m^3 with error bars of 11 to 12%. Our result are comparable to the values observed on Sept. 2 but higher than the last data by factors of 3 to 4. Considering the difference in observation conditions etc., we should say our result is roughly consistent to the observations of CTBT station in Takasaki as well as the observations of Fukushima University.

Inhalation dose is calculated by multiplying the amount of intake and conversion coefficients. The former is the product of activity concentration, breathing rate and time. If we use the largest value (type 'S') of the ICRP conversion coefficients for adults[(6)], committed effective doses due to 1-day inhalation are 2.72×10^{-10} Sv and 7.18×10^{-10} Sv for ^{134}Cs and ^{137}Cs respectively. If the conversion coefficients of ECRR[(7)] are employed, value for ^{137}Cs rises to 1.2×10^{-9} Sv. Breathing rate of 22.21m^3/day is used throughout the above calculation.

5. Discussion

Even for full 1-year breathing of this contaminated air, above doses are very small compared to the dose due to gamma-rays from the radioactive materials deposited on the earth's surface or on buildings. However, recent decontamination works may cause a risk of increasing the air contamination. Worst of all, the accident has not terminated yet and no one knows what happens next. Therefore a continuous and systematic monitoring of the radioactivity in the atmosphere will be necessary.

The inhalation dose may be significant in the early days after 3.11 all over Fukushima Prefecture. Air dusts of this period are kept on the enormous number of air filters carried by cars which were then on the roads. Among them there will be a number of filters which started or stopped to work during the days of dense radioactive plume. If one can compare the activities of filters which began or stopped aspiration at different times, we could possibly obtain time-differential values of radioactivity concentration. Collection of these filters is desirable with their records of time-profile of use.

We express thanks to Mr. R. Doi at Radioisotope Institute for Basic and Clinical Medicine, Kurume University, for granting to use the Ge detector. We also thank Prof. A. Watanabe at Fukushima University for sending their recent data on atmospheric concentration of radioactive Cs measured at his campus.

References

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