

Technical preparations for atmospheric radioactivity monitoring

Yutaka Miyamoto,
Tetsuzo Oda,
Takeo Adachi,
Hiroshi Noguchi,
Hideo Nishimura,
Shigekazu Usuda

Abstract To evaluate the suitability of a location for a radionuclide monitoring station of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) at Takasaki, Japan, the assessment was done in terms of the concentration of natural background radioactive nuclides in airborne dust and soil samples. The samples were taken four times at the proposed site and several points around the site, then their radioactivities were measured by gamma-ray spectrometry. For the airborne dust samples, only natural background radionuclides: ^{212}Pb , ^7Be and ^{214}Pb were detected. The radioactivity concentrations of these radionuclides varied in four sampling periods but were almost the same as the previous measurements. For the soil samples, the concentration of an anthropogenic radionuclide, ^{137}Cs , fluctuated among the different sampling points and periods, but the concentration of ^{40}K was constant. These concentrations were 2 to 3 times lower than those of the soil taken at JAERI Tokai. Based on these results, the proposed site was evaluated and has been accepted by the CTBT Organization as a CTBT radionuclide monitoring station.

Key words airborne dust • background activity • CTBT • environmental radioactivity • nuclear explosion • radionuclide monitoring

Introduction

In order to verify compliance with Comprehensive Nuclear-Test-Ban Treaty (CTBT), a cost effective International Monitoring System (IMS) is to be established, including four monitoring techniques: radionuclide, seism, infrasound and hydroacoustics. The IMS using radionuclide monitoring techniques consists of 80 stations in the world.

Radioactive particles will be collected, at each of 80 stations, from the atmosphere onto an air-filter with a high-volume air sampler at a minimum flow rate of $500\text{ m}^3/\text{h}$, and the noble gases at 40 of the 80 stations will also be collected into a charcoal trap [7]. Then, their radioactivities will be measured either automatically or manually by a separate gamma-ray spectrometric system.

The Protocol of CTBT specifies two radionuclide monitoring stations in Japan should be established at Takasaki City (for both particle and the noble gases monitoring) and Okinawa Prefecture (for particle monitoring) [7].

The JAERI has to be working for the establishment of these radionuclide monitoring stations as a responsible organization in Japan [4]. For establishing the monitoring stations, it is important to avoid an undesirable place, e.g. existence of high natural and/or anthropogenic radioactive sources, bad support infrastructures, unfavorable meteorological conditions such as wind impact and upper airflow coupling. Some site survey works have been performed by

Y. Miyamoto[✉], T. Oda, T. Adachi, H. Noguchi,
H. Nishimura, S. Usuda
Japan Atomic Energy Research Institute (JAERI),
2-4, Shirakata-shirane, Tokai, Ibaraki 319-1195, Japan,
Tel.: +81 29/ 282 5518, Fax: +81 29/ 282 6950,
e-mail: miyamoto@sghsun.tokai.jaeri.go.jp

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the JAERI at the JAERI-Takasaki location to evaluate its suitability for the establishment of an IMS station. The preparatory commission of the CTBT Organization decides the suitability for an IMS station based on its criteria and the site survey results. For the assessment of existence of high radioactivity sources, airborne dust and surface soil samples were taken at the proposed site, then analyzed by gamma-ray spectrometry with a High-Purity Germanium (HP-Ge) detector. This paper focuses on the background radioactivity of airborne dust and surface soil samples at the proposed site in Takasaki (JAERI-Takasaki).

Methods

The airborne dust samples were collected on a glass fiber filter (ADVANTEC Inc., GB-100R) with a high-volume sampler (SIBATA Scientific Technology Ltd., HV-1000N) at a flow rate of 60 m³/h for a period of 11 days in maximum. The nominal collection efficiency of the filter is 99.99% at particles of 0.3 μm in diameter. After the sampling, the filter was packed into a cylindrical plastic container (60 mm in diameter, 22 mm in height). Then, the radioactivity of the airborne dust was measured with an HP-Ge detector (relative efficiency of 30% at 1.33 MeV) for 80 000 seconds at JAERI-Tokai. The measurements were repeated twice or three times to accurately determine the radioactivity. The radionuclide concentrations, except for ²¹⁴Pb, were corrected for radioactive decay during the periods of sampling, preparation and measurement. The decay correction of ²¹⁴Pb concentrations was done only for the periods of preparation and measurement, because uncertainty of the correction for the sampling period was very large.

Surface soil samples (each about 2000 g) were collected from the central point of the site, 4 points at a distance of 5 m north, south, east and west, and 2 points at a distance of 100 m east and west from the center of the site. The dimension of soil sample for this measurement was an approximately 5 cm depth and a 20 cm square. The samples were oven-dried at 110°C for 4–7 days. The dried samples were sieved to eliminate grains of more than 2 mm in diameter. Each sample aliquot (260–350 g) was packed into a cylindrical plastic container (95 mm in diameter, 58 mm in height). Then, the radioactivity of the samples was measured with the same detector as that used for measuring the airborne dusts.

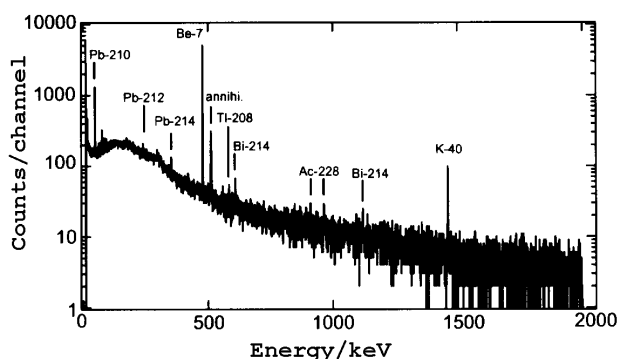


Fig. 1. Typical gamma-ray spectrum of airborne dust. The airborne dust was taken at the site proposed for the CTBT Takasaki station from August 27 to September 7, 1998 (11 days). The total flow volume was 15 843 m³. The airborne dust was measured with an HP-Ge detector for 80 000 seconds after 4.9 days of airborne-taking.

The counting efficiency of the HP-Ge detector was determined with two calibrated standard sources which have the same geometry as each dust and soil sample. The radioactivity of sources already had been certified by Laboratoire de Mesure des Rayonnements Ionisants (LMRI), whose present name is CERCA Co.

The airborne dust and soil samples were taken four times at the proposed site; November 1997, February 1998, May 1998 and September 1998. The background radioactivities were compared with those measured at the JAERI-Tokai in November 1997 and in February 1998.

Results and discussion

While several natural background nuclides were detected, anthropogenic radionuclides, such as ¹³⁷Cs and ¹⁴⁰Ba were not detected in the airborne dust samples beyond the detection limit of 0.004 mBq/m³ and 0.05 mBq/m³, respectively. Fig. 1 shows a typical gamma-ray spectrum obtained from the measurements. The concentrations of representative radionuclides detected in the airborne dust samples are shown in Fig. 2. The concentration of natural background radionuclides fluctuated among the samples taken in each period. The concentration of ²¹²Pb in the sample taken in November was high (105 mBq/m³), and that in May was low (22 mBq/m³). In contrast with the fluctuation pattern of concentration of ²¹²Pb, the concentration of ⁷Be in the sample taken in November was low (2.5 mBq/m³), and those taken in February and May were high (5 and 4 mBq/m³, respectively). Both the concentration ranges of ²¹²Pb (22–105 mBq/m³) and ⁷Be (2.5–5.4 mBq/m³) were almost the same as the previously reported values of 40–120 mBq/m³ for ²¹²Pb [8] and of 2–7 mBq/m³ for ⁷Be [1].

Lead-212 is originated from ²³²Th in soils, and ⁷Be is originating from reactions of cosmic-ray and air. These radionuclides are captured on airborne dust, and carried by the wind. Therefore, these fluctuations may be caused by seasonal changes of the climate. At the site, in January, prevailing wind direction was north-northwest, and the monthly precipitation has its minimum throughout the year. The precipitation increases at the approach to August when the prevailing wind direction changes to east-southeast.

In the soil samples, several natural background radionuclides and an anthropogenic radionuclide, i.e. ¹³⁷Cs, were detected. Fig. 3 shows the concentration of radionuclides in the soil samples taken at 7 positions in each sampling period. It was found that the soil sampled at the center of the proposed site contained the highest amounts of ¹³⁷Cs (45–70 Bq/kg in dry soil) among the soils taken at the positions. The soil sampled at 100 m west from the center contained the lowest one (14–29 Bq/kg). The ¹³⁷Cs concentrations in each sampling period were not uniform, and the deviation was more than 50%. The ¹³⁷Cs concentration of soil taken at the JAERI Tokai (25–110 Bq/kg) was fluctuated among the concentrations of the soils taken at seven positions in the JAERI-Takasaki. The ²¹²Pb concentration varied in the range of 10–14 Bq/kg with the difference of sampling positions and periods. The average concentration of ⁴⁰K in these samples was 190 Bq/kg (±4.6%, 1-sigma). The concentrations of ²¹²Pb and ⁴⁰K sampled at the JAERI-Tokai

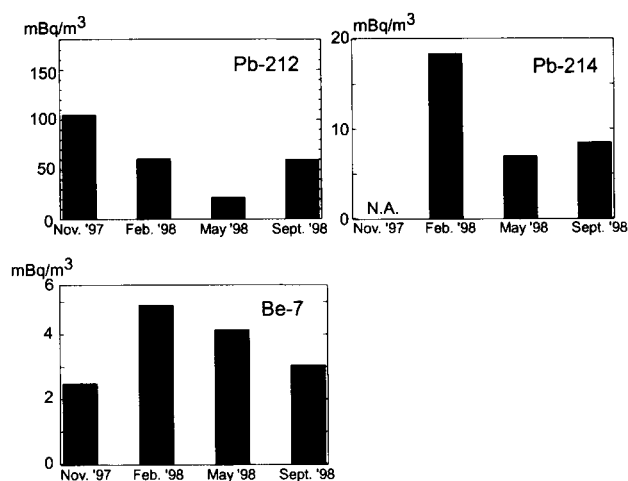


Fig. 2. Radioactivity concentrations of ^{212}Pb , ^{214}Pb and ^7Be in the airborne dusts taken at the site proposed for the CTBT Takasaki station. "N.A." means that the data was not analysed due to the long cooling time.

were 22 and 500 Bq/kg, respectively. The concentrations at the proposed site were 2 to 3 times lower than those at the JAERI-Tokai. The concentrations of ^{137}Cs and ^{40}K in the soil taken at more than 45 points in Japan are compiled in some papers [5, 9]. The concentrations of these two radionuclides in the papers were as follows; ^{137}Cs : 0.48–79 Bq/kg, (average 54 ± 0.8 Bq/kg), ^{40}K : 0.72–2.65% as metallic potassium, which is equivalent to 217–800 Bq/kg of ^{40}K . The concentration results of these two radionuclides in this work were within the variation of the reported values.

From the results of radioactivity concentration in the airborne dust and soil samples taken at the proposed site, it is concluded that there is no undesirable anthropogenic radionuclide to carry out the atmospheric radioactivity monitoring and that the concentrations of natural background radionuclides are not exceptionally high in Japan.

The site which we proposed for the IMS radionuclide monitoring station at Takasaki has been accepted as the suitable one by the preparatory commission for the CTBT Organization in 1998, and the construction of the station is under way.

The site survey work, on the site which we proposed for the IMS radionuclide monitoring station at Okinawa was finished and the proposed site for the CTBT radionuclide station was accepted in March 2001.

Conclusion

The following results were found by measuring the background radioactivity of the airborne dust and the soil at the site proposed for the Takasaki CTBT radionuclide monitoring station.

In the airborne dust samples, the ^{212}Pb concentration range were 22–105 mBq/m^3 , and the range of ^7Be was 2.5–5.4 mBq/m^3 . The ^{212}Pb concentration was high in November and the fluctuation pattern of ^7Be concentration was in contrast with that of ^{212}Pb . Neither ^{137}Cs nor ^{140}Ba were detected in the airborne dusts.

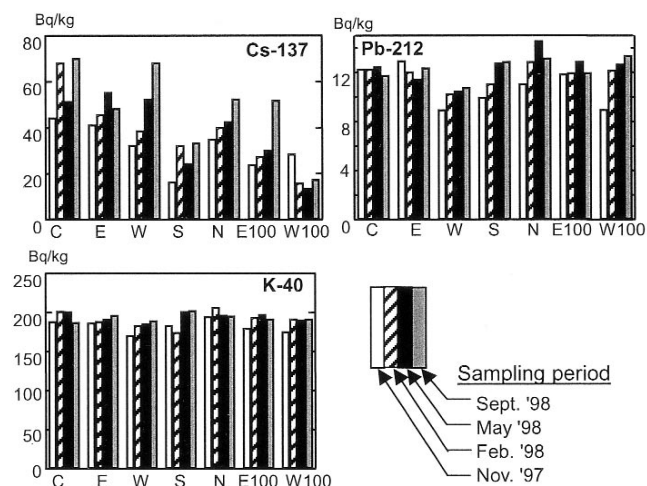


Fig. 3. Radioactivity concentrations of ^{137}Cs , ^{212}Pb and ^{40}K in the soil samples taken at the site proposed for the CTBT Takasaki station. The unit of radioactivity concentration is Bq/kg in dry soil. The initial letters on the horizontal axis mean sampling point from the proposed position; C: Center of the position, E: 5 m east, W: 5 m west, S: 5 m south, N: 5 m north, E100: 100 m east, W100: 100 m west.

In the soil samples, the concentration of ^{137}Cs and ^{212}Pb were fluctuated among the different seven sampling points and four periods. The concentration range of ^{137}Cs and ^{212}Pb were 14–70 Bq/kg and 10–14 Bq/kg, respectively. The concentration of ^{40}K was constant, 190 Bq/kg ($\pm 4.6\%$, 1-sigma).

Since, no undesirable radioactivity was detected at the proposed Takasaki-site, this site has been accepted as the suitable place for the CTBT station.

The continuously observed and accumulated data at the CTBT monitoring stations may be also important not only for the CTBT purpose but also for the monitoring of radioactivities caused by a nuclear power accident and in the field of environmental science. These daily monitorings are very useful if the investigator's interests relate to the variation of radionuclide concentrations with the locality (latitude) and season [2, 3, 6].

For example, ^7Be is one of the useful tracers for study on airflow behavior. From the results of ^7Be concentration, measured at several test-running CTBT radionuclide stations, it tends to show the following seasonal variation: in the Northern Hemisphere, the ^7Be concentrations in May to August are high, but in the Southern Hemisphere the concentrations in those periods are low. If the daily monitoring data at the 80 CTBT stations and meteorological data will be obtained, the relation between the seasonal variation and airflows can be more clarified.

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