Journal of Environmental Radioactivity 111 (2012) 33–37

Contents lists available at SciVerse ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Monitoring of aerosols in Tsukuba after Fukushima Nuclear Power Plant incident in 2011

Yutaka Kanai*

Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Higashi, Tsukuba, Ibaraki 305-8567, Japan

ARTICLE INFO

Article history:
Received 18 July 2011
Received in revised form
12 October 2011
Accepted 14 October 2011
Available online 8 November 2011

Keywords:
Artificial nuclides
Aerosol
Tsukuba
^{137}Cs
^{134}Cs
Sedimentology

ABSTRACT

Artificial radionuclides were released into the atmosphere by the Fukushima Dai-ichi Nuclear Power Plant incident after a strong earthquake on 11 March 2011. Aerosol monitoring at the Geological Survey of Japan, Tsukuba, was started 20 d after the incident. Radionuclides such as ^{99}Mo/^{99m}Tc, ^{132}Te/^{132}I, ^{129m}Te/^{129}Te, ^{131}I, ^{137}Cs, ^{136}Cs, ^{134}Cs, ^{140}Ba/^{140}La, ^{110m}Ag, and ^{95}Nb were observed and, with the exception of ^{137}Cs and ^{134}Cs, these radionuclides decreased to below the limit of detection in the middle of June. The activity ratio of atmospheric ^{134}Cs/^{137}Cs in aerosols decreased over time almost following physical decays. Therefore, the ^{134}Cs/^{137}Cs activity ratio in the averaged air mass in this study could be regarded as homogeneous although those of several reactors in the Nuclear Power Plant were not ascertained. A further research on the released ^{137}Cs and ^{134}Cs would be necessary for the sedimentology of lake sediment.

1. Introduction

On 11 March 2011, a strong earthquake occurred in eastern Japan and the subsequent large-scale tsunami damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), resulting in venting operation, suppression pool damage, and hydrogen explosion. Thus, large amounts of radioactive nuclides were released from the reactor containers into the environment. The main nuclides exhausted were ^{131}I, ^{137}Cs, and ^{134}Cs (IAEA, 2011; TEPCO, 2011). The amounts of ^{131}I and ^{137}Cs were estimated to be about 1.6×10^{17} Bq and 1.5×10^{16} Bq, respectively (Kantei, 2011).

The activities of these nuclides in the atmosphere were important information to assess the radiation dose of the public and reported by many observatory stations around the world (CTBTO, 2011; Manolopoulou et al., 2011; Pittauerova et al., 2011; Leon et al., 2011; Bolsunovsky and Dementyev, 2011). We also began to monitor them in Tsukuba, about 170 km south of the FDNPP (Fig. 1). Although the activity of ^{131}I decreased because of its relatively short half-life (8 d), those of ^{137}Cs and ^{134}Cs remained detectable.

On the other hand, radionuclides in the environment, such as ^{210}Pb and ^{137}Cs, are used for the dating of lake and sea bottom sediments in the field of sedimentology (Appleby and Oldfield, 1978; Cochran et al., 1998; Goldberg and Koide, 1962; Koide et al., 1972, 1973; Krishnaswami et al., 1971). We have studied the sedimentary environments of lake sediments by measuring activities of ^{210}Pb and ^{137}Cs (for example, Kanai and Ikehara, 1995; Kanai et al., 1997, 2002). Lead-210 is the descendant nuclide of gaseous ^{222}Rn, and attaches to aerosols to produce excess in atmospheric deposition. The excess ^{210}Pb in the sediment decreases with time after sedimentation, and the sedimentation rate is calculated by the slope between the logarithm of excess ^{210}Pb and sediment depth. On the other hand, ^{137}Cs is used as a marker for the year 1954 when its activity was first detected and for 1963 when its activity in the atmosphere and sediment reached maximum levels (Peirson, 1971).

The radioactivities in aerosols are also important for sedimentological studies as they contribute to the dating of sediments. In this study, data of radionuclide concentrations in the aerosols observed at the Geological Survey of Japan (GSJ) in Tsukuba were examined.

2. Experimental

2.1. Samples

Air sampling was started on 31 March 2001. The method used for sampling of aerosols was the same as that described previously (Kanai et al., 2003, 2005). The particulate (not gaseous) radionuclides in the aerosols were collected with a high volume air sampler (HV-1000F, Shibata Scientific Co., Ltd.) equipped with a polyflon

* Tel.: +81 29 861 3865; fax: +81 29 861 3643.
E-mail address: y.kanai@aist.go.jp.

0265-931X/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2011.10.011

Fig. 1. Map of sampling site.

filter (PF040, about 25 cm × 20 cm × 0.95 mm thick, capture efficiency of 95% for 0.3 μm particle, Advantec Co., Ltd.). It was set on the rooftop of a building 14 m above ground level at the GSJ (140.13°E, 36.06°N) in Tsukuba. Its flow rate was maintained at 1000 l/min. Aerosols were collected on the filter for about 6 h in the early runs, and then for 3 or 4 d. The sampling data are shown in Appendix A. The filter was folded into 1/16 size, put into a sealed plastic container (78 mm phi × 46 mm high), and its activity was measured.

2.2. Apparatus for measurement

A gamma ray spectrometer with a well-type Ge detector (GWL-120-16-LB-AWT-HJ-S, SEIKO EG&G ORTEC) was used for measurement. The relative efficiency of the Ge detector was 20%, and its well was 16 mm in diameter and 40 mm in depth. The shielding of the Ge detector was composed of 115 mm Pb, 48 mm Fe, 2 mm Cd, 20 mm oxygen-free Cu, and 5 mm acryl lining. The details of the equipment were reported elsewhere (Kanai and Saito, in press). The detection efficiency was determined by measuring the NBL (New Brunswick Laboratory) reference materials (the analyzed sample No. 79, 1% Th ore, and the counter calibration sample, 1% U ore). The NBL counter calibration sample (1% U ore) was diluted with SiO_2 and 5% Fe_2O_3 into 0.5% U to match the matrix for the correction of self absorption because the aerosols in Japan contained several percent of Fe_2O_3 (Ohta et al., 2003). The analyzed sample No. 79 and the diluted counter calibration sample were prepared with the same geometry as the filter sample (folded into 1/16 size) and were in equilibrium within the short-lived daughter nuclides of Rn. The correction of sum effect was conducted by measuring ^{134}Cs activity about 15 cm above the Ge detector (Komura, 2006).

The filter in a sealed plastic container was put on the Ge detector and measured for about 1 d. The spectra were analyzed using Gamma Studio software (DS-P240/W32, SEIKO EG&G) and the activities were corrected at the time when aerosol sampling started.

3. Results and discussion

3.1. Radionuclide concentrations in aerosols at the GSJ

Many observation stations reported the daily radiation dose and the nuclide activities (e.g. CTBTO, 2011). TEPCO (2011) also reported the daily activities of radionuclides in the air within the FDNPP. Although they are observed continuously and show daily variation, the detection limit is relatively high because the air sampling time is only 10 or 20 min. For example, the detection limit of ^{137}Cs is $2–4 \times 10^{-7}$ Bq/cm^3 (0.2–0.4 Bq/m^3). In Tsukuba, KEK (2011) has also observed daily variation of air concentrations since 15 March. In our study, the sampling of aerosol started from 31 March and the sampling time was relatively long (3 or 4 d) to obtain averaged data and more precise data (the average detection limit of ^{137}Cs was 8×10^{-5} Bq/m^3) although the time resolution of daily variation decreased.

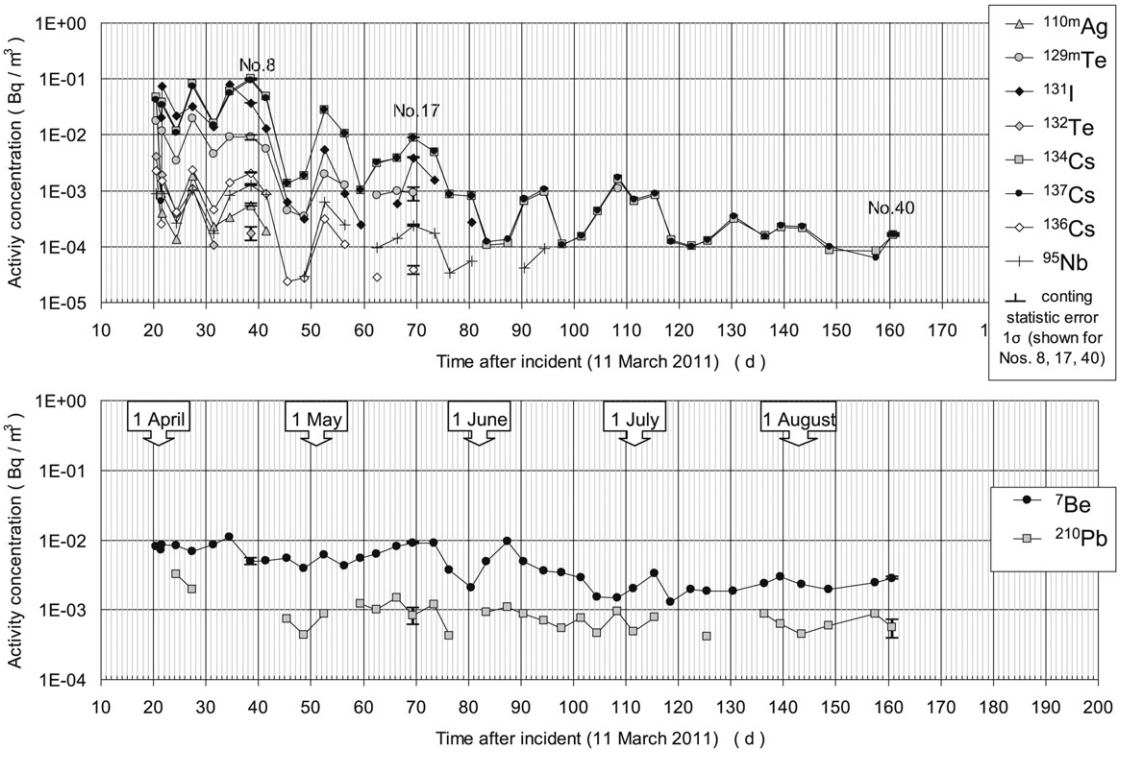

Fig. 2. Activity concentrations of radionuclides in aerosols observed at the GSJ in 2011. Counting statistic error bars are shown for sample Nos. 8, 17, and 40 to show typical errors.