Journal of Nuclear and Radiochemical Sciences, Vol. 8, No. 2, pp. 149-153, 2007

High Resolution Measurements of Temporal Variations of Airborne ^{210}Po, ^{210}Pb, and ^{7}Be at Ishikawa Prefecture, Facing the Sea of Japan

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Received: February 1, 2007; In Final Form: July 14, 2007

High-resolution measurements of temporal variations of airborne ^{210}Pb and ^{7}Be were performed by ultralow background γ-spectrometry at Ogoya Underground Laboratory (OUL). Moreover, high-resolution measurements of ^{210}Po were also made by α-spectrometry at Low Level Radioactivity Laboratory (LLRL). In this paper, the results focusing mainly on cyclic variation with ~1 week period observed in winter season, and the high resolution measurements conducted at the time of approach of typhoon and Kosa events are reported.

1. Introduction

Natural and artificial radionuclides have been used as versatile tracers in geochemical studies. The concentrations of these nuclides are generally very low, and therefore a sufficient amount of samples or a longer counting period is required to obtain reliable data. However, by the use of ultralow background Ge detectors at Ogoya Underground Laboratory (OUL), it is possible to detect extremely low levels of environmental radionuclides.

The radioactivity levels of airborne ^{210}Pb (half-life: 22.3 y) and cosmic-ray-induced ^{7}Be (half-life: 53.3 d) in the surface air are of the order of ~mBq m^{-3}, which is three orders of magnitude lower than that of airborne ^{222}Rn (half-life: 3.8 d). The concentrations of these nuclides in the air are expected to vary on a time scale comparable to that of ^{222}Rn, but no precise measurements have been performed due to the difficulty in the detection of low levels of ^{210}Pb and ^{7}Be without using extremely-high volume air samplers or extremely-high sensitive detection techniques. By using ultralow background Ge detectors at the OUL (270 m.w.e.), it is possible to detect mBq levels of ^{210}Pb and ^{7}Be,[1,2] which correspond to only 10 min of sampling time by an ordinary high-volume air sampler. In this study, we have continuously monitored the concentration of airborne ^{210}Po (half-life: 138.4 d), ^{210}Pb, and ^{7}Be at the Low Level Radioactivity Laboratory (LLRL) in Kanazawa University, Nomi City; we changed filter paper of air sampler at intervals of 2–6 hours or 1 day and investigated the correlations between temporal variations of these nuclides and meteorological conditions.

Polonium-210 is daughter product of ^{222}Rn and ^{210}Pb, and has been used to estimate residence time of airborne particles,[3] to understand air mass transportation, and to evaluate the influence of volcanic activities.[4–6] The concentration of airborne ^{210}Po increases due to forest fires, transport of continental dust by wind, intrusion of older air masses from the stratosphere or the high troposphere,[7] etc. Measurements of airborne ^{210}Po have been performed to investigate the usefulness of this nuclide as a geochemical tracer from the viewpoints mentioned above.

In this study, daily variations and high resolution measurements (2–6 hour intervals) of temporal variations of airborne ^{210}Po, ^{210}Pb, and ^{7}Be during the approach of typhoons and Kosa events at LLRL in Ishikawa Prefecture, which faces the Sea of Japan, are reported. The locations of LLRL and OUL are shown in Figure 1.

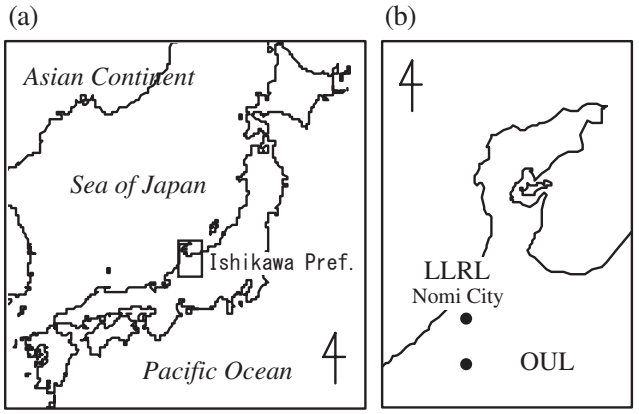

Figure 1. Location maps of (a) Ishikawa Prefecture, Japan and (b) sampling point (LLRL) and Ogoya Underground Laboratory (OUL); they are located around 136–138°E and 36–37.5°N.

2. Experimental

2.1. Sampling. We have been monitoring and analyzing airborne ^{210}Po, ^{210}Pb, and ^{7}Be at LLRL almost every day since November 17, 2005. In addition, during meteorological events such as the approach of typhoons and Kosa events, 2–6 hour short-interval samplings were performed to investigate the effects of the events.

Airborne particles were collected on a silica-fiber filter (ADVANTEC QR-100, 254 mm × 203 mm) using a high-volume air sampler (Sibata HV-1000F) at a flow rate of 700–900 L min^{-1} on the roof (10 m a.g.l, and 50 m a.s.l) of LLRL. In order to measure low-level ^{210}Pb concentrations, the blank levels of natural radionuclides in the filter papers should be sufficiently low to minimize the correction to be made in the analysis. The blank levels of the uranium and thorium series nuclides and ^{40}K in an ordinary glass-fiber filter (ADVANTEC GW-100) and a silica-fiber filter (ADVANTEC QR-100) were measured using two and four filters, respectively.[8] As seen in Table 1 and Figure 2, it is apparent that the levels of natural radionuclides in the silica-fiber filter are found to be orders of magnitude lower than those in the glass-fiber filter. Thus, the silica-fiber filter was chosen for the sampling in this work. The

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Published on Web 10/19/2007

TABLE 1: Blank levels of natural radionuclides of a glass-fiber and a silica-fiber filter

Nuclide	mBq/filter		Silica/Glass Ratio
	Glass	Silica	
^{210}Pb	122	8.00	0.07
^{238}U	150	13.0	0.09
^{226}Ra	81.7	7.67	0.09
^{228}Ra	65.0	1.67	0.03
^{228}Th	53.3	1.17	0.02
^{40}K	4990	16.7	<0.01

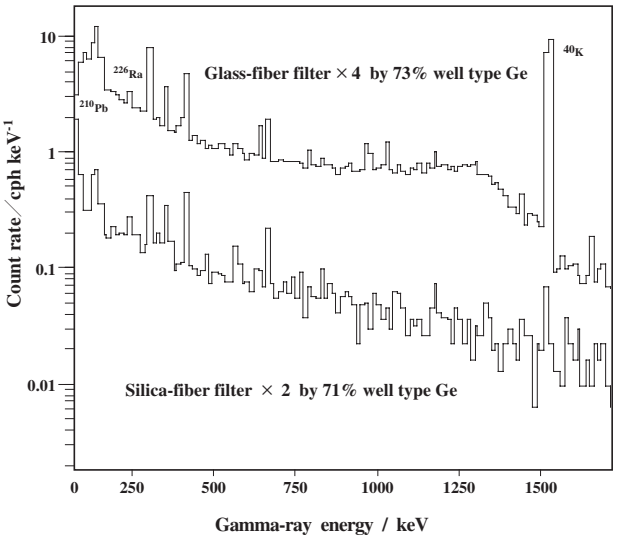

Figure 2. Comparison of the radioactivity levels of a glass-fiber filter and a silica-fiber filter.

^{210}Pb radioactivity in the filter was subtracted from the measured ^{210}Pb radioactivity. The ^{210}Po radioactivity in the filter is expected to be nearly in radioactive equilibrium with its parent nuclide ^{210}Pb. However, the contribution of ^{210}Po in the filter to the measured ^{210}Po radioactivity was found to be negligibly low, probably due to the fact that ^{210}Po was firmly held within the silica fiber and it was not easily extracted even by 8 M HNO_3 used in chemical treatment described in sec. 2.2.2.

2.2. Measurements and data analyses.

2.2.1 Radioactivities of ^{210}Pb and ^{7}Be. Airborne ^{210}Pb and ^{7}Be that were collected on the filters were measured nondestructively by a large-volume ultralow background planar-type and/or well-type Ge detectors at OUL.[9] The efficiency of the detectors was calibrated using mock-up standards prepared from a mixture of the reference material No. 42-1 (4.04% uranium in radioactive equilibrium issued from New Brunswick Laboratory (NBL, USDOE)) and a pulverized silica-fiber filter.

In order to measure γ-rays of ^{210}Pb and ^{7}Be, 78% of the aliquot in the filter was compressed into a disk with a diameter of 35 mm using a hydraulic press to prepare a counting source. The measurements were carried out for one to several days after the disintegration of the short-lived nuclides ^{214}Pb and ^{212}Pb. The ^{210}Pb and ^{7}Be radioactivities were determined by employing the peak counts of 46.5 keV (I_γ = 4.05%) and 477.8 keV (I_γ = 10.59%), respectively, and were corrected for radioactive decay. The concentrations of these radioactivities (Bq m^{-3}) were calculated on the assumption that they were constant during each sampling interval.

2.2.2 Radioactivity of ^{210}Po. In order to measure the radioactivity of ^{210}Po collected on the filters, radiochemical separations were performed within one week (several days) after

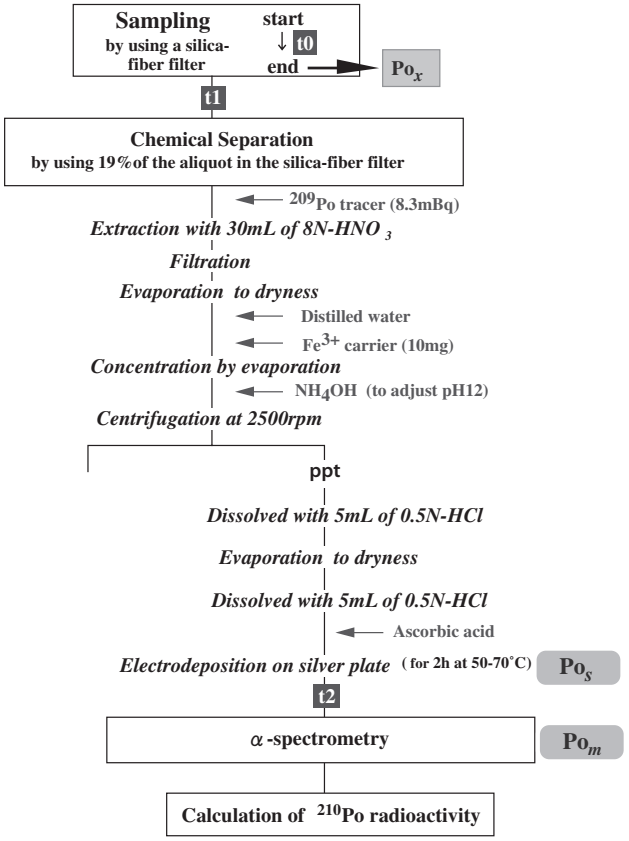

Figure 3. Procedure for the radiochemical analysis of ^{210}Po.

sampling to suppress the ingrowth of ^{210}Po from ^{210}Pb and ^{210}Bi (half-life: 5.0 d).

As depicted in Figure 3, ^{210}Po was extracted from 19% of the aliquot in the silica-fiber filter by heating the filter at 250 °C with 30 mL of 8 M HNO_3 and using ^{209}Po (8.3 mBq) as a yield tracer. The extract was filtrated through an ordinary filter paper and was concentrated at 250 °C. Equal amounts of distilled water and the concentrated HNO_3 solution were mixed along with 10 mg of Fe^{3+} ions which was added as a carrier. Subsequently, polonium (^{210}Po and ^{209}Po tracer) was coprecipitated with $Fe(OH)_3$ at pH 12 which was maintained by using NH_4OH. The $Fe(OH)_3$ precipitate was collected after centrifuging it at 2500 rpm and redissolved in 5 mL of 0.5 N HCl, and then dried up again. The residue was dissolved in 5 mL of 0.5 N HCl, and a sufficient amount of ascorbic acid was added to clear the solution by masking Fe^{3+}. A 0.2-mm thick silver plate with a size of 10 × 10 mm was immersed in the sample solution for 2 hours at 50–70 °C for electrochemically depositing polonium on it. The polonium radioactivity deposited on the silver plate was measured at LLRL by α-spectrometry using a surface-barrier-type Si detector.

The concentration of ^{210}Po was calculated by the following process. The ^{210}Po radioactivity at the time of measurement (Po_m) was first calculated from the radioactivity of spiked ^{209}Po and the ^{210}Po/^{209}Po counts ratio. Then, the ^{210}Po radioactivity during the chemical separation (Po_s) was calculated by correcting for radioactive decay for the time interval between the electrodeposition and the measurement ($t2$). Next, the ^{210}Po radioactivity at the end of the sampling (Po_x) was calculated by subtracting the contribution of ^{210}Po derived from ^{210}Pb and ^{210}Bi during the time between the end of sampling and the separation ($t1$). The ^{210}Po concentration in the atmosphere was finally calculated by considering the growth and decay of ^{210}Pb, ^{210}Bi, and ^{210}Po during sampling ($t0$), although, in this study, the magnitude of this effect was negligibly low because $t0$ was less than one day.

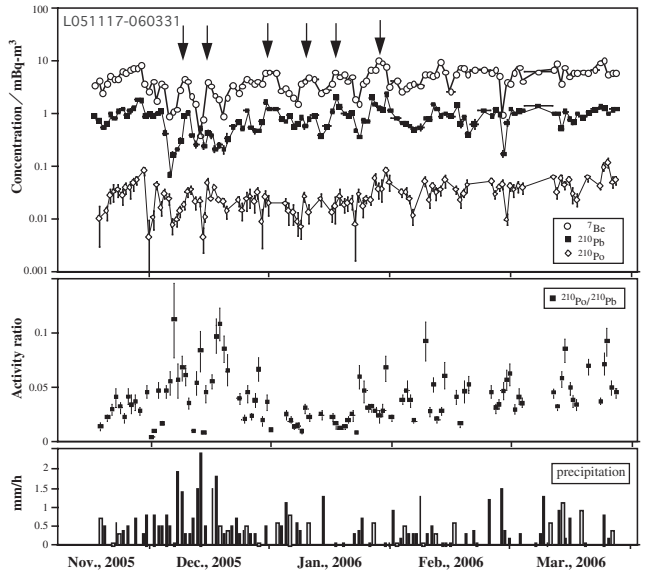

Figure 4. Daily variations of airborne ^{7}Be, ^{210}Pb, and ^{210}Po concentrations, and ^{210}Po/^{210}Pb radioactivity ratios at LLRL from November 17, 2005 to March 31, 2006.

3. Results and Discussions

3.1. Daily variations of airborne radionuclides. Figure 4 shows the variations of ^{7}Be, ^{210}Pb, and ^{210}Po measured daily from November 17 in 2005 to March in 2006 together with daily precipitation. Measured period was only 5 months, however, general variation patterns of ^{210}Pb and ^{7}Be were found to be very similar; lowest in December and gradual increase toward March. In the previous measurements[10] (May to December, 2003) conducted simultaneously at LLRL, Hegura Island (located 50 km from Wajima), and Shishiku Plateau (10 km East of LLRL), seasonal variation patterns of ^{210}Pb and ^{7}Be were found to be almost the same; decrease from spring (highest value at May) to summer (minimum in July–August), then gradual increase toward November and decrease again in winter (December–January). The lack of previous measurement could be covered by the present study.

In the present study, characteristic variations of about a week of cycle was first recognized both for ^{7}Be and ^{210}Pb in December and January. As pointed by arrows, peak concentrations of ^{7}Be and ^{210}Pb in each cycle were observed mostly just after the occurrence of precipitation (snow fall), therefore, the valley shaped decrease of ^{7}Be and ^{210}Pb radioactivities may be explained by being scavenged due to snow fall.

With regard to ^{210}Po, general trend in its variation pattern is similar to that of the parent nuclide ^{210}Pb, because both nuclides are long-lived daughter nuclides of ^{222}Rn. This similarity is often obscured on days when rain and/or snow fall, which shorten apparent residence time of airborne particles. As known from Figure 4, high ^{210}Po/^{210}Pb radioactivity ratio was observed mostly at the time when ^{210}Pb concentration showed its minimum in each cycle. Although not confirmed yet, high ^{210}Po/^{210}Pb values over 0.1 may be explained by the effect of fine particles, which may act as the nucleus of snowflake, derived from the resuspension of the surface soil containing high ^{210}Po/^{210}Pb radioactivity ratio (nearly in equilibrium).

3.2. Variations of radioactivities during the approach of typhoon. Figure 5 shows the variations of the ^{7}Be, ^{210}Pb, and ^{210}Po concentrations and the ^{210}Po/^{210}Pb radioactivity ratio during the approach and passage of typhoon No. 14 on September 5–9, 2005. The concentrations of ^{210}Pb and ^{7}Be showed a decrease when the typhoon passed over the Sea of Japan on September 7.

The concentrations of ^{210}Pb and ^{7}Be were measured to be in the ranges of 0.053–0.349 and 0.141–2.23 mBq m^{-3}, respectively, and the minimum values during the typhoon period were approximately 1/20–1/10 of the corresponding values in calm weather. These minimum concentrations were observed at a time (approximately 9:00 on September 7) close to the passage of the typhoon. Since significant rainfall was not observed at this time, the predominant removal process for airborne radionuclides was the inflow of the air masses containing low-concentration ^{7}Be and ^{210}Pb, and not due to washout by rainfall.

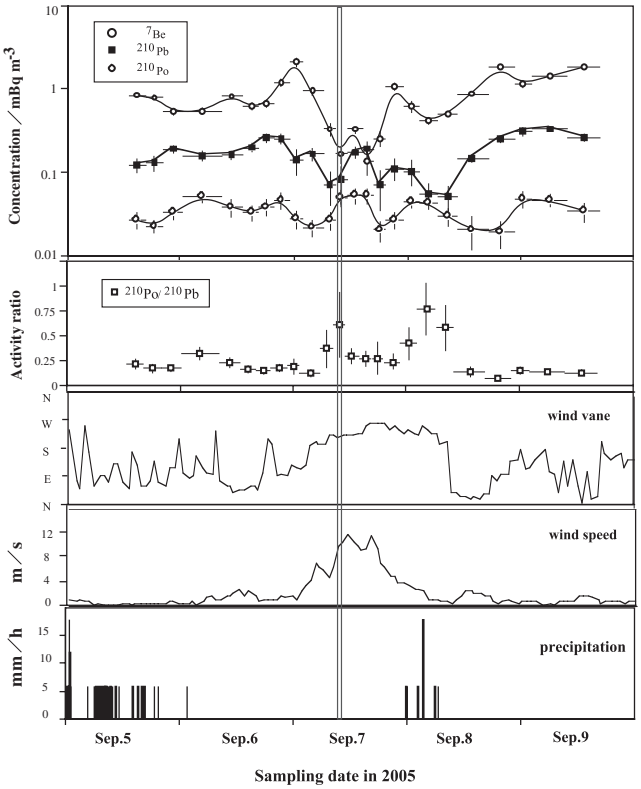

Figure 5. Variations of airborne ^{7}Be, ^{210}Pb, and ^{210}Po concentrations, and ^{210}Po/^{210}Pb radioactivity ratios during the approach and/or passage of a typhoon (September 5–9, 2005). The meteorology data were supplied by the Environment Radiation Department, Ishikawa Prefectural Institute of Public Health and Environmental Science.

A typhoon is a tropical cyclone originating from the low-latitude Pacific Ocean. The concentration of airborne ^{7}Be is low in the low-latitude area. Moreover, the production rate of ^{210}Pb is very low on the sea because of the extremely low emanation of parent ^{222}Rn from the ocean surface.[11–14] Around the typhoon, strong winds raise sea-salt particles showing high ^{210}Po/^{210}Pb radioactivity ratio (approximately 0.6)[15] from the ocean surface.[16,17] The sea-salt particles proceed northward with the air masses containing low-concentration ^{7}Be due to strong winds. As a result, it is considered that the ^{210}Pb and ^{7}Be concentrations became low and the ^{210}Po/^{210}Pb radioactivity ratio showed high during the typhoon period.

On the other hand, the concentration of cosmogenic ^{7}Be showed a drastic decrease (1/20–1/10) when compared with that of ^{210}Pb. Consequently, the radioactivity ratio ^{210}Pb /^{7}Be showed a considerably high value (1.43 ± 0.58). Abe et al.[9] reported that this kind of variation was also observed in the case of the passage of cold fronts. These observations may be explained under the following assumptions.

Both the nuclides ^{210}Pb and ^{7}Be attached to aerosol particles were once scavenged by the washout process, but the inflow (recovering) rates of these nuclides are different due to the difference in their origin. The inflow of ^{7}Be from the upper atmosphere is slow as compared with that of ^{210}Pb originating from airborne ^{222}Rn near the ground surface.

3.3. Variations of radioactivities during Kosa events. At least three Kosa events were observed in April 2006 (April 8,

18–19, and 24–25), and sampling was performed at approximately 2–6 hour intervals around the Kosa events and at 1 day intervals for the other days. As observed in Figure 6, the concentrations of all three radionuclides ^{7}Be, ^{210}Pb, and ^{210}Po showed increases in the 2nd and 3rd events, and in all the events ^{210}Po/^{210}Pb radioactivity ratios showed higher values (max: 0.28) compared with those in other days. The amount of airborne particles showed extremely high values, however, no correlation was observed between the amount of particles and concentrations of these nuclides. On the other hand, the concentrations of ^{7}Be and ^{210}Pb showed decrease in the 1st event (April 8). These phenomena may be explained as follows.

Kosa particles originating from arid zones are raised up to 2–5 km above the ground[18] by convection, carried over the Japanese islands by strong westerlies, and descend gravitationally and/or by downward current of air mass to the ground surface. On the other hand, the concentrations of cosmogenic ^{7}Be produced in the stratosphere and upper troposphere,[19] and ^{210}Pb in the stratosphere are greater in value as compared to the values in the upper troposphere.[20,21] It is well known that the concentrations of airborne ^{7}Be and ^{210}Pb shows high values in Japan during the spring season[23] due to the active inflow of air mass from the stratosphere (which shows a high ^{210}Po/^{210}Pb radioactivity ratio)[15] to the upper troposphere in mid-latitude regions.[21,23,24] Abe et al.[9] suggested that the active inflow of air mass is one of the reasons for the high ^{210}Pb and ^{7}Be concentrations during Kosa events; the high concentrations should not be due to radionuclides being attached to Kosa particles and their being washed out together, because no correlation was observed between the concentrations of Kosa particles and radionuclides. The inflow flux of upper aerosol particles to the lower atmosphere is independent of the concentration of Kosa particles; hence, the mass concentration of aerosol particles is not proportional to the ^{7}Be and ^{210}Pb concentrations. The decrease of ^{7}Be and ^{210}Pb concentrations in the 1st event may be explained by the influence of rainfall.

Abe et al.[9] reported that anthropogenic ^{137}Cs was detected in some of samples collected during Kosa events. Contrary to the cases of ^{7}Be and ^{210}Pb, the relationship between ^{137}Cs and the mass concentration of airborne dust showed a linear correlation, which indicates that the ^{137}Cs collected during Kosa events originates from those attached to and transported together with Kosa particles.

The suggested reasons for the high ^{210}Po/^{210}Pb radioactivity ratios during Kosa events are the active inflow of air containing high ^{210}Po/^{210}Pb values, and also the high concentration of Kosa particles in which nuclides from ^{222}Rn to ^{210}Po in uranium decay series can be assumed to be in radioactive equilibrium (^{210}Po/^{210}Pb = 1.0).[7] However, all of ^{210}Po in Kosa particles cannot completely be extracted by radiochemical extraction using 8 M HNO_3 (sec. 2.2.2). This may be the reason why clear correlation between the increase of the weight of Kosa particles and that of ^{210}Po/^{210}Pb radioactivity ratios were not observed.

It is considered that the measurements of the size distribution of ^{7}Be, ^{210}Pb, and ^{210}Po is profitable in order to know if Kosa particles have an efficiency as a scavenger of aerosol particles.

4. Conclusion

Daily variations and highly time-resolution analysis (2–6 hours) of airborne ^{210}Po, ^{210}Pb, and ^{7}Be have been investigated at Ishikawa Prefecture, which faces the Sea of Japan.

When large-scale meteorological changes occurred, as in the case of the typhoons and Kosa events, characteristic variation patterns were observed in the ^{210}Pb and ^{7}Be concentrations. High resolution measurements of temporal variations of the airborne radionuclides proved to be very useful in investigating the geochemical behaviors of airborne particles during meteorological variations resulting from air mass movements.

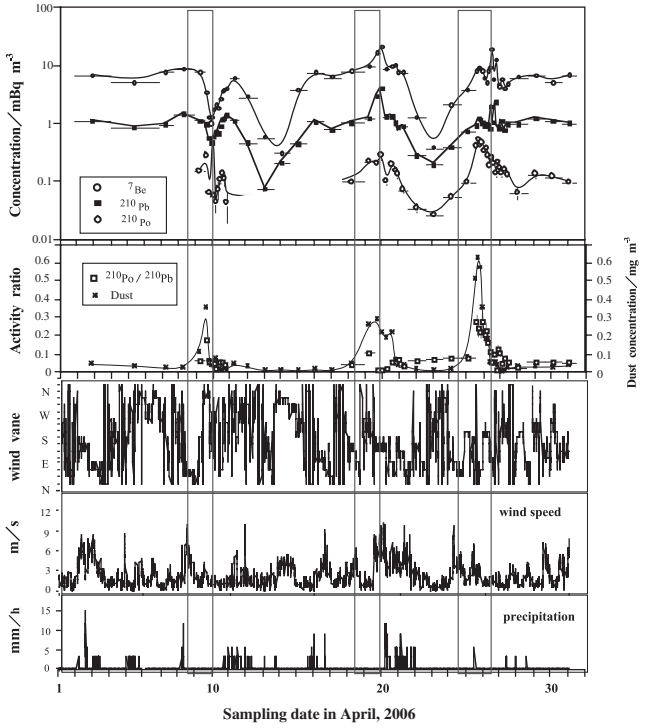

Figure 6. Effect of the arrival of Kosa events on airborne ^{7}Be, ^{210}Pb, and ^{210}Po concentrations, and ^{210}Po/^{210}Pb radioactivity ratios (April, 2006). The meteorology data were supplied by the Environment Radiation Department, Ishikawa Prefectural Institute of Public Health and Environmental Science.

With regard to ^{210}Po, the general trend in its variation patterns was similar to that of ^{210}Pb, which is the parent nuclide of ^{210}Po. However, characteristic variation in the ^{210}Po/^{210}Pb radioactivity ratio was observed to be depending on the weather condition.

Acknowledgment. This study was performed as a part of the Kanazawa University 21st-Century COE Program "Environmental Monitoring and Prediction of Long- and Short-term Dynamics of Pan-Japan Sea Area."

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