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Highly Time-Resolved Measurements of Airborne Radionuclides by Extremely Low Background γ-ray Spectrometry: Their Variations by Typical Meteorological Events

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In order to investigate variation of concentrations of airborne radionuclides, ^{214}Pb, ^{212}Pb, ^{7}Be, and ^{210}Pb, accompanied with a typical meteorological event, such as passage of cold front, approach of typhoon, and Asian dust (Kosa), highly time-resolved (2–6 hours) measurements of natural radionuclides were performed by ultra low background γ-ray spectrometry. Surface air concentrations of these nuclides showed characteristic temporal changes by each meteorological phenomenon. In addition, variation patterns were found to be obviously different between short-lived ^{214}Pb and ^{212}Pb and long-lived ^{7}Be and ^{210}Pb. One of causes of the difference might be explained by difference of their sources: local for short-lived nuclides and regional for long-lived ones.

1. Introduction

Most of the radionuclides in the atmosphere are attached to aerosol particles and behave along with these particles. Therefore, they have been considered to be useful tracers for the analyses of various atmospheric processes. In this reason, observations of dynamic state of atmospheric radionuclides have been performed to investigate the processes such as transport and/or mixing of air masses, which are characterized by residence times of aerosol particles in the atmosphere, etc. A number of investigations have been made for explaining regional atmospheric processes with the use of atmospheric concentrations and/or deposition fluxes of ^{210}Pb (half-life: 22.3 y), which is a long-lived decay product of ^{222}Rn emanating from ground surface, and ^{7}Be (53.3 d) produced by the nuclear spallation reaction between secondary cosmic rays and nuclei in the upper atmosphere.[1-6] On the other hand, clarifications of advection and diffusion processes and removal processes of aerosol particles attaching the nuclides from the atmosphere have been attempted from variability analyses of short-lived ^{214}Pb and ^{212}Pb which are decay products of airborne ^{222}Rn and ^{220}Rn, respectively.[7-11]

The behavior of aerosols depends largely on meteorological factors of ambient air, conditions of which change sensitively with atmospheric phenomena such as Kosa events and others. In order to use airborne radionuclides attaching to aerosol particles as tracers, it is important to perform observations with time intervals suitable to time scale of atmospheric phenomena of interest. In humid climate regions, wet deposition accompanied with rainfall or snowfall is a dominant removal process of aerosols from atmosphere and it often occurs in a few hours order. That is, observation with short time intervals is desirable to discuss the removal processes such as precipitation scavenging. Additionally, observations on various radionuclides with different particular life-times and sources simultaneously would provide various time and spatial information on weather changes.

Ogoya Underground Laboratory (OUL) of Low Level Radioactivity Laboratory (LLRL), Kanazawa University, was opened in a tunnel of former Ogoya copper mine with rock covering of 135 m (270 m.w.e.) in 1995. Background levels of 16 Ge detectors equipped in OUL, which are shielded with thick rock cover and lead manufactured more than 200 years ago, are about 2 orders of magnitude lower than ordinary low background Ge detectors in ground-level laboratories. Gamma-ray spectrometry using 16 Ge detectors at OUL allows to measure long-lived ^{7}Be and ^{210}Pb along with short-lived ^{214}Pb and ^{212}Pb with higher time resolution (0.5–3 hours, corresponding to only 30-180 m^3 of air volume).[12,13] In this paper, we report concentration variations of these nuclides during the times of typical weather change events described above.

2. Experimental

2.1. Sampling. Sampling of aerosol particles was performed on the roof (10 m above ground level, and 50 m above sea level) of LLRL, Kanazawa University in Nomi, Ishikawa Prefecture, Japan. Location map is shown in Figure 1. Aerosol particles were collected on a weighed filter using a high volume air sampler (SIBATA HV-1000F) at a flow rate of 900 L min^{-1} with 2–6 hours of sampling intervals which were varied depending on the atmospheric condition on each occasion. Silica fiber filter (ADVANTEC QR-100, size: 203 mm × 254 mm) was chosen for sampling because concentrations of naturally occurring radionuclides (^{210}Pb: 1.2 mBq g^{-1}, ^{40}K: 5.3 mBq g^{-1}) were found to be much lower than those in glass fiber filter (GB-100R-810A).[14] Sampling was made during the time of (1) passage of cold front (2004/12/18 and 20 Japan Standard Time: J.S.T.), (2) approach of typhoon (2005/9/7 J.S.T.), and (3) Kosa event (2006/4/8, 18-19, and 24-25 J.S.T.). Referring information from the local meteorological observatory, highly time-resolved observations (samplings) had been carried out during a few

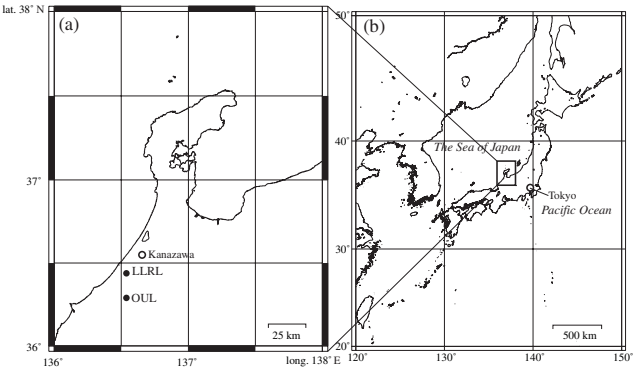

Figure 1. Location map of the sampling point and the OUL, with overview of near Japan.

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days before and a few days after the each event in the case of (1) and (2). However, at the Kosa events, samplings were started at the time when the event was confirmed visually.

2.2. Gamma-ray measurements. In order to measure short-lived ^{214}Pb and ^{212}Pb, γ-ray counting source was prepared immediately after the sampling as follows. After few minutes of drying under an infrared lamp and weighing (except period (1)), 77% aliquot of the filter was compressed into a disk with 35 mm in diameter and 3 mm in thickness using hydraulic press. Gamma-ray measurements of ^{214}Pb and ^{212}Pb were performed by using 2 coaxial-type Ge detectors installed in LLRL for 2–12 hours. In order to measure long-lived ^{7}Be and ^{210}Pb, after disintegration of short-lived nuclides, remeasurements were performed for 2–5 days by using ultra low background planer- and well-type Ge detectors at OUL.

2.3. Data analyses. Peak counts of 46.5, 239, and 478 keV γ-rays were used to determine ^{210}Pb, ^{212}Pb, and ^{7}Be, respectively. On the other hand, summation of the count at 242, 295, and 352 keV γ-rays was used for ^{214}Pb. Detection efficiencies of each detector were calibrated by the measurements of standard sources prepared from the mixture of reference material No. 42-1 (4.04% uranium) issued from New Brunswick Laboratory (NBL), USDOE and silica fiber filter as a host material.

Activity of an airborne radionuclide a (Bq m^{-3}) at the sampling was calculated by the following formula:

$$a = \frac{C\lambda^2}{(1 - e^{-\lambda t_1})(e^{-\lambda t_2} - e^{-\lambda t_3})F\varepsilon B}$$

where C, λ, t_1, t_2, and t_3 are the area of peak counts during the counting time, the disintegration constant (s^{-1}), the time of air sampling (s), the time difference between the end of sampling time and the start time of measurement (s), and the time difference between the end of sampling time and the end time of measurement (s), respectively. And F, ε, and B denote the flow rate of air sampler (m^3 s^{-1}), the detection efficiency of detector at the energy of γ-ray from the corresponding nuclide, and the branching ratio of γ-ray emission, respectively. This formula implies that the concentration a is an average value during each sampling interval.

The average wind speed and precipitation data at every 10 min intervals of on-site and surface weather chart around Japan were obtained from Ishikawa Prefectural Institute of Public Health and Environmental Science and from Meteorological Agency of Japan (http://www.jma.go.jp/jma/indexe.html), respectively.

Ordinary observations made at sampling intervals of 1–2 day(s) for the other study were performed continuously since 2003. Parts of the data from these observations (not published yet) were appropriately used for comparison with present study.

3. Results and discussion

3.1. Variation of concentrations of airborne radionuclides during the passage of cold front. During period from December 17 to 22, 2004, when two cold fronts passed over this area, highly time-resolved sampling was performed to investigate the effect of the passage of cold front event. Variations of ^{214}Pb, ^{212}Pb, ^{7}Be, and ^{210}Pb concentrations, wind speed, and precipitation are plotted in Figure 2. During this period, the concentrations of all nuclides measured were varying more than an order of magnitude; 1.47±0.04–14.1±0.2 Bq m^{-3} for ^{214}Pb, 9.28±0.78–83.2±6.5 mBq m^{-3} for ^{212}Pb, 0.149±0.034–9.71±1.00 mBq m^{-3} for ^{7}Be and 0.105±0.023–3.13±0.24 mBq m^{-3} for ^{210}Pb.

According to surface weather conditions, two cold fronts had passed over the sampling location around 9 p.m. on Dec. 18 J.S.T. and around 6 p.m. on Dec. 20 J.S.T., respectively. As can be seen in Figure 2, all of the nuclides showed clear decreases just at the times of the passages of cold fronts. This decrease might be explained mainly by washout due to rainfall. It is noteworthy that the decreases of long-lived nuclides at the passages of cold fronts were more predominant than those of short-lived ones. This may be explained that the activities of short-lived nuclides are rapidly recovered by the decays of their gaseous precursor radon isotopes, which are not removed by precipitation. On the other hand, supplies of long-lived ^{7}Be and ^{210}Pb in the surface air require longer times. As another explanation, differences of efficiencies of washout due to sizes of aerosol particles containing the radionuclides might be considered. According to Papastefanou,[15,16] sizes of aerosol particles (activity median aerodynamic diameter: AMAD) attaching long-lived ^{7}Be and ^{210}Pb tend to be larger than those attaching short-lived ^{214}Pb and ^{212}Pb; i.e. ^{214}Pb: 0.07–0.21, ^{212}Pb: 0.09–0.23, ^{7}Be: 0.29–0.50, and ^{210}Pb: 0.28–0.49 μm. On the other hand, as reported by Hirose et al,[17] a correlation between washout ratios (meaning efficiencies of washout) and particle diameters for aerosol particles with submicrometer size is not so clear. Therefore, differences of efficiencies of washout between long-lived ^{7}Be and ^{210}Pb and short-lived ^{214}Pb and ^{212}Pb may be caused by differences of supply rates between them.

It is also noted that the concentrations of ^{7}Be and ^{210}Pb showed an increasing tendency along with the approach of cold front. A cold front transported over Japan area in winter season is provided by proceeding in the east direction of cold air mass which originates with Siberian High pushing up warm air mass over the Sea of Japan. Yamamoto et al[18] observed remarkably high monthly deposition in the locations of the Japan Sea side in winter season, and explained as follows. An inversion layer, which inhibits diffusion of ^{222}Rn and its short-lived progeny to upper atmosphere, is formed by radiation

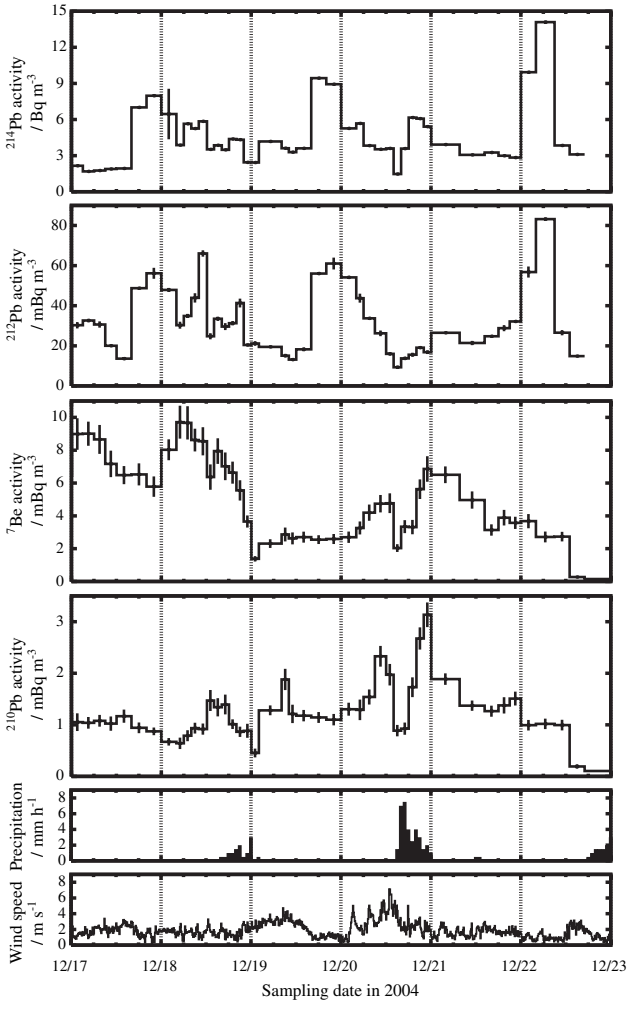

Figure 2. Variation of airborne radionuclides, precipitation and wind speed during the event of passages of cold fronts on Dec. 19–20, 2004.

cooling in Asian continent. As a consequence, cold air mass originating from the continent in winter contains higher ^{210}Pb activity than in the other seasons. Additionally, a cold air mass also comes down from arctic region of high latitude where production rate of ^{7}Be is high to Japan region in the same season. Therefore, convection cloud developed by transport of the cold air masses causes rainfall and/or snowfall containing high ^{7}Be and ^{210}Pb concentrations over the Japan Sea side in winter season. The increases of atmospheric concentrations of these nuclides during the approach of the cold front observed in this study may also be explained by the same mechanism.

Rapid decrease of the long-lived nuclides in the afternoon of Dec. 22 is considered to be caused by washout due to snowfall which started at that time. In this connection, studies[19,20] suggested that washout effect of snowfall is greater than that of rainfall because snow particles concentrate more aerosol particles due to their larger surface areas and longer flight duration than raindrops.

3.2. Variation of concentrations during the approach of Typhoon. In order to know effects of a typhoon on the surface air concentrations of the nuclides, highly time-resolved observation was performed during the period of September 5 to 9, 2005. On Sep. 7, typhoon No. 14 passed over the Sea of Japan. The closest approach of the typhoon was at about 9 a.m. J.S.T. when it was proceeding due northeast located about 200 km northwest of the sampling location. Figure 3 shows a surface weather chart at that time and a track of the typhoon. From the pressure distribution, it can be confirmed that the sampling location was involved in effective area of the typhoon. Variations of the nuclide concentrations, mass concentration of airborne dust, wind speed and precipitation are shown in Figure 4. The concentrations of ^{214}Pb, ^{212}Pb, ^{7}Be, and ^{210}Pb were measured to be in the ranges of 0.272±0.014–10.4±0.1 Bq m^{-3}, 7.97±0.81–131±10 mBq m^{-3}, 0.141±0.048–2.23±0.26 mBq m^{-3}, and 0.053±0.017–0.349±0.030 mBq m^{-3}, respectively, and the minimum values during the typhoon period were approximately 1/20–1/10 of the corresponding values obtained in calm weather in ordinary observations. On the contrary, the higher mass concentrations of airborne dust than usual level of 20–40 μg m^{-3} were observed along with the approach of the typhoon. A maximum value of 110 μg m^{-3} was observed just after the time of the closest approach of the typhoon. This increase may be explained not by the contribution of soil components stirred up by strong wind, but by sea salt particles generated massively on the ocean because of no increase of natural uranium and thorium series nuclides, ^{40}K, and artificial ^{137}Cs which are of typical soil particle origin.

Since occurrence of minimum concentrations of all the nuclides coincided with the closest time of typhoon, unusual decreases of their concentrations may be attributed to effect of the typhoon. The decreases of concentrations of the nuclides are caused by the fact that the typhoon originates from low latitudes Pacific Ocean. There is a report [21] which showed that low concentrations of ^{7}Be in surface air at low latitudes region is explained by low production rate of ^{7}Be at upper atmosphere due to low geomagnetic latitude and low inflow of upper atmosphere to the lower atmosphere due to updraft of the atmospheric general circulation. In addition, on the ocean, not only concentrations of short-lived ^{214}Pb and ^{212}Pb but also that of long-lived ^{210}Pb is low[6] due to little dissipation of precursor radon isotopes from seawater. Moreover, most of the aerosol particles are scavenged by the rain during the transport of typhoon and, consequently, radionuclides attached to aerosol particles are effectively removed from air mass carried by typhoon. Because a typhoon transports such maritime air mass with low nuclide activities, it seems reasonable that the concentrations of long-lived ^{7}Be and ^{210}Pb had decreased since few days before the closest time of the typhoon. On the other hand, the decreases of the concentrations of short-lived ^{214}Pb and ^{212}Pb began just before the closest time when strong wind started to blow. Because the origins of the short-lived nuclides are neighborhood region, their concentrations hardly decrease only by transport of maritime air mass without strong wind. The drastic decreases of the short-lived nuclides are considered to be caused not only by horizontal mixing with maritime air mass but also by vertical mixing with upper atmosphere, in which concentrations of precursor radon isotopes are low, due to turbulence caused by strong wind.

It is noteworthy that ^{210}Pb/^{7}Be activity ratio varied more than 20 times from minimum value of 0.065±0.024 to maximum value of 1.43±0.58 during the approach of the typhoon.

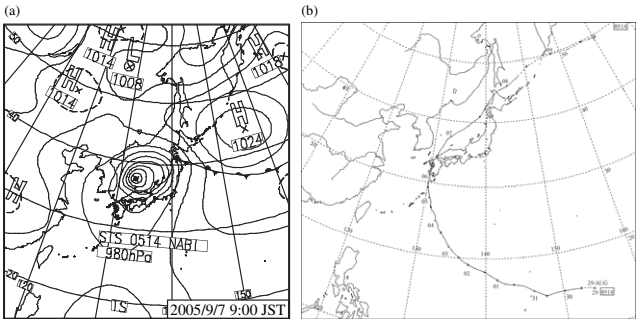

Figure 3. (a) The surface whether chart at the time of the closest approach of the typhoon No. 14 in 2005 and (b) the tracks of the typhoon. (Diverted from the website Meteorological Agency, Japan, http://www.data.kishou.go.jp/yohou/kaisetu/hibiten/index.html).

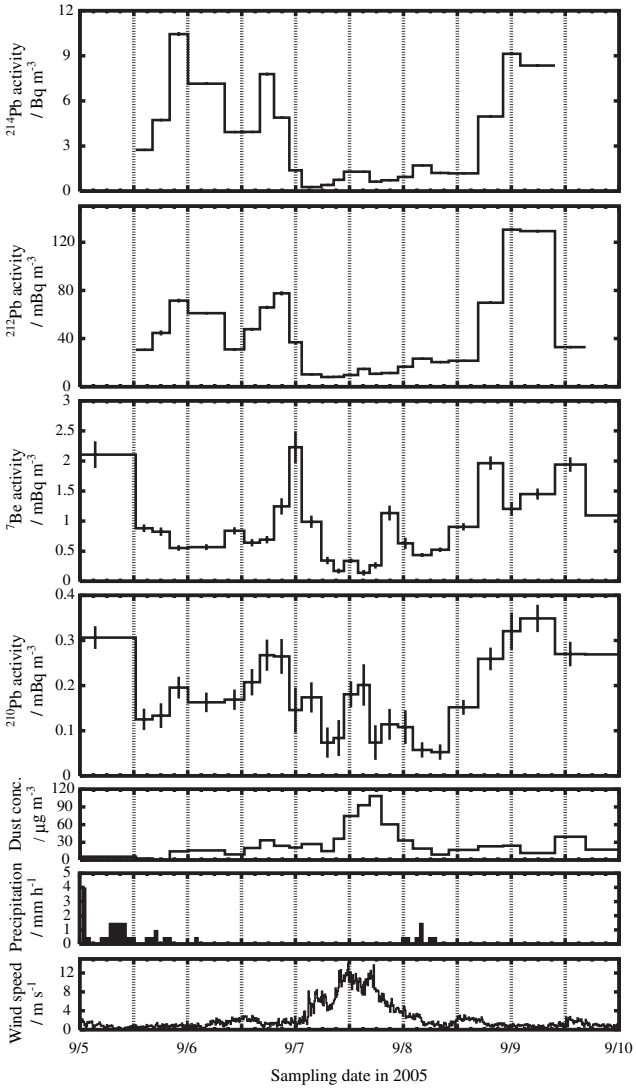

Figure 4. Variation of airborne radionuclides, dust concentration, precipitation and wind speed during the event of an approach of a typhoon on Sep. 7–8, 2005.

Therefore, there was no positive correlation between ^{7}Be and ^{210}Pb during this period, although positive correlation has generally been seen in ordinary observations.[2,4,22,23] Such a large fluctuation of the ^{210}Pb/^{7}Be ratios was also observed at the time of passage of cold front described above. This kind of variation may be explained by a hypothesis that although both nuclides attached on aerosol particles are once scavenged by washout process, flow-in rates of these nuclides are different due to the difference of their origin, i.e. ^{7}Be from upper atmosphere and ^{210}Pb from land surface.

3.3. Variation of concentrations during Kosa event. Three Kosa events were observed at the sampling location in April 2006. Variations of the concentrations of the airborne radionuclides during these periods are shown in Figure 5 together with mass concentrations of airborne dust and meteorological data. Drastic increase of the mass concentration up to 630 μg m^{-3} (value of sampling for about 3 hours), which is more than 10 times higher than ordinary values of 20–40 μg m^{-3}, was observed during the Kosa event.

Short-lived radionuclides, ^{214}Pb and ^{212}Pb, showed essentially the oft-reported diurnal variation,[8,11,24] i.e. the concentrations of these nuclides are high at first hours of morning and low at evening and decrease due to strong wind and/or precipitation. Kosa particles were transported from the Asian continent to over Japan area for 2 days at the shortest case. Since the traveling time is 4–5 times longer than the half-life of ^{212}Pb, we expect a little effect of ^{212}Pb originating from the continent. It is also difficult to distinguish from predominant component occurring from the local area. The effect of ^{212}Pb derived from a long range transport may be observed if sampling is performed at higher altitude, over the sea or other locations where the concentration of ^{212}Pb is expected to be extremely low.

As can be seen in Figure 5, the concentrations of long-lived ^{7}Be and ^{210}Pb were high during the Kosa events in April. Relationships between the concentrations of ^{7}Be and ^{210}Pb and that of airborne dust are given in Figure 6 along with data of ordinary daily observation in April. Figure 6 suggests that the concentrations of both ^{7}Be and ^{210}Pb tend to be high according to the increase of the dust concentrations in surface air although there seems to be no linear relationship between radionuclides and dust concentrations. This can be explained as follows. It is well known that concentrations of both ^{7}Be and ^{210}Pb in surface air are high in Japan in spring seasons.[2,4] This is explained by the inflow of air from stratosphere containing rather high amounts of cosmogenic ^{7}Be and terrigenous ^{210}Pb to upper troposphere due to folding of tropopause at mid-latitude region in spring season.[5,25] On the other hand, Kosa particles are blown up from land surface to upper atmosphere up to altitude of several thousands meters at dry regions, where are main origins of Kosa particles, due to upward current accompanied with strong storm. And then, Kosa particles are transported by westerlies and deposited on ground surface by gravitational settling and/or downward current. Therefore, it is considered that aerosol particles attaching ^{7}Be and ^{210}Pb descend from the upper troposphere when Kosa phenomena are observed. However, since the sources of Kosa particles and the aerosol particles attaching the long-lived nuclides are different, there may not be linear relationship between them. As a result described above, it is

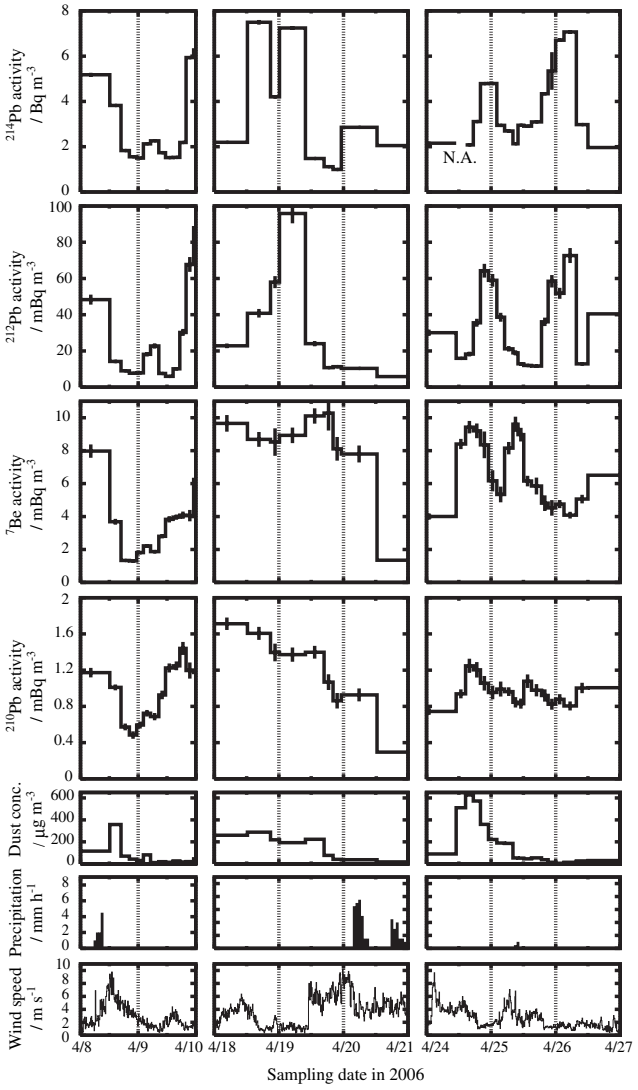

Figure 5. Variation of airborne radionuclides, dust concentration, precipitation and wind speed during the event of influxes of Kosa on Apr. 8, 18–19 and 24–25, 2006.

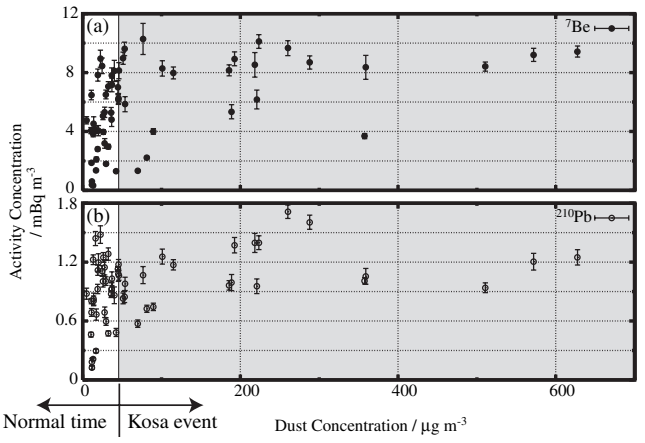

Figure 6. Relationships between atmospheric concentrations of (a) ^{7}Be and (b) ^{210}Pb and dust concentration in Apr. 2006 including Kosa events.

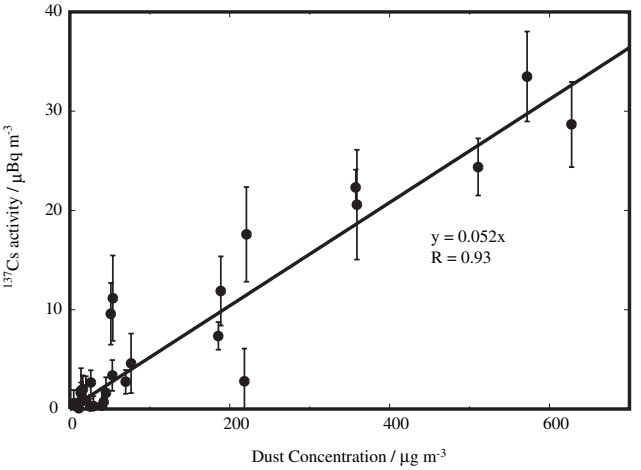

Figure 7. Relationship between atmospheric concentration of ^{137}Cs and dust concentration in Apr. 2006 including Kosa events.

considered that rather high concentrations of ^{7}Be and ^{210}Pb in surface air were observed at the time of Kosa events.

It is also noted that anthropogenic ^{137}Cs were detected in some of samples collected during Kosa events. Relationship between the ^{137}Cs and mass concentration of airborne dust is shown in Figure 7. As is clear in this figure, a linear correlation can be seen different from the cases of ^{7}Be or ^{210}Pb mentioned above. There is no source of ^{137}Cs in the atmosphere including stratosphere. The ^{137}Cs deposited on land surface is considered to be transported together with Kosa particles due to re-suspension process. From this result, a specific activity of ^{137}Cs in Kosa particles obtained during the Kosa events is estimated to be 52 mBq g^{-1}.

4. Conclusion

Highly time-resolved measurements of the surface air concentrations of ^{214}Pb, ^{212}Pb, ^{7}Be, and ^{210}Pb (+^{137}Cs) were performed during the period of typical weather change events, passage of cold front, approach of typhoon, and Kosa events. It is noteworthy that variation patterns obviously differed between short-lived ^{214}Pb and ^{212}Pb and long-lived ^{7}Be and ^{210}Pb. The short-lived nuclides showed connections with wind speed which is corresponding to a local meteorological element. In contrast, concentration variations of long-lived nuclides are influenced by larger scale meteorological phenomena. The difference is explained mainly by the difference between origins of short-lived and long-lived nuclides, namely, local and regional ones, respectively. Therefore, highly time-resolved measurements of long-lived nuclides in surface air are very useful to know sources and histories of air masses including marked phenomena.

Moreover, differences between long-lived nuclides, whose causes are assumed to be due to difference of their origins, could be seen. This study is qualitative currently, but it is expected to be quantitative by advance in further researches.

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References

(1) K. Hirose, T. Honda, S. Yagishita, Y. Igarashi, and M. Aoyama, Atmos. Environ. **38**, 6601 (2004).
(2) J. Sato, T. Doi, T. Segawa, and S. Sugawara, Geochem. J. **28**, 123 (1994).
(3) D. Todorovic, D. Popovic, G. Djuric, and M. Radenkovic, J. Environ. Radioactivity **79**, 297 (2005).
(4) M. Yoshimori, Adv. Space Res. **36**, 828 (2005).
(5) W. M. Burton and N. G. Stewart, Nature **186**, 584 (1960).
(6) K. Yokota and S. Tsunogai, Geochem. **25**, 59 (1991) (in Japanese).
(7) A. A. Ahmed, A. Mohamed, A. E. Ali, A. Barakat, M. A. El-Hady, and A. El-Hussein, J. Environ. Radioactivity **77**, 275 (2004).
(8) G. M. Marcazzan, E. Caprioli, G. Valli, and R. Vecchi, J. Environ. Radioactivity **65**, 77 (2003).
(9) H. Kojima, J. Aerosol Res. Jpn. **10**, 265 (1995) (in Japanese).
(10) H. W. Gäggeler, D. T. Jost, U. Baltensperger, and M. Schwikowski, Atmos. Environ. **29**, 607 (1995).
(11) Y. Koike, K. Kukita, T. Nakamura, and J. Sato, Radioisotopes **54**, 87 (2005).
(12) K. Komura, *Proc. 1997 Intern. Symp. Environmental Radiation*, eds. T. Tsujimoto and Y. Ogawa, Tsuruga, Fukui, Japan (1997) pa11.
(13) K. Komura and Y. Hamajima, Appl. Radiat. Isot. **61**, 185 (2004).
(14) Y. Yamaguchi, K. Komura, Y. Murata, M.N. Manikandan, T. Iida, J. Moriizumi, and S.Hirao, *Proc. Fifth Workshop on Environmental Radioactivity*, ed. T. Miura, KEK, Tsukuba, Japan (2004) p137 (in Japanese).
(15) C. Papastefanou and E. A. Bondietti, Sci. Total Environ. **70**, 69 (1988).
(16) C. Papastefanou, *Proc. Third Intern. Summer School*, Low-level Measurements of Radioactivity in the Environment, eds. M. García-León and R. García-Tenorío, Huelva, Spain (1993) p249.
(17) K. Hirose, S. Takatani, and M. Aoyama, J. Atmos. Chem. **17**, 61 (1993).
(18) M. Yamamoto, A. Sakaguchi, K. Sasaki, K. Hirose, Y. Igarashi, and C. K. Kim, J. Environ. Radioactivity **86**, 110 (2006).
(19) M. Murakami, T. Kimura, C. Magono, and K. Kikuchi, J. Meteor. Soc. Jpn. **61**, 346 (1983).
(20) Y. Ishikawa and H. Murakami, J. Environ. Radioactivity **26**, 19 (1995).
(21) H. Nagai, J. Aerosol Res. Jpn. **15**, 321 (2000) (in Japanese).
(22) M. Baskaran and G. E. Shaw, Aerosol Sci. **32**, 443 (2001).
(23) A. S. Likuku, J. Environ. Radioactivity **87**, 289 (2006).
(24) D. F. Measday and E. C. Y. Ho, Nucl. Instrum. Methods Phys. Res. B **213**, 464 (2004).
(25) L. Kownacka, Nukleonika **47**, 79 (2002).