Article

Atmospheric Concentrations of ^{210}Pb and ^7Be Observed in Okinawa Islands

Akira TANAHARA*, Fumiya NAKAEMA**, Yoshitaka ZAMAMI** and Takemitsu ARAKAKI***

*Instrumental Research Center, University of the Ryukyus

**Graduate School of Engineering and Science, University of the Ryukyus

***Department of Chemistry, Biology and Marine Science, Faculty of Science, University of the Ryukyus
1 Senbaru, Nishihara-cho, Nakagami-gun, Okinawa Pref. 903-0213, Japan

Received July 16, 2013

Atmospheric measurements of naturally occurring radionuclides ^{210}Pb and ^7Be were conducted at ground level in the Okinawa Islands. Weekly data collected from Okinawa-jima over a seven-year period from 2004 to 2011 and Kume-jima and Minamidaito-jima over a two-year period from 2008 to 2010 are presented and discussed. The monthly averaged activity concentrations of ^{210}Pb at three sites showed a "one-peak" seasonal variation — the maximum peak appeared from autumn to spring — and ^7Be showed a pattern similar to that of ^{210}Pb . Activity concentrations of ^{210}Pb and ^7Be at Okinawa Islands ranged from 0.06 to 1.98 mBq/m³ with a mean of 0.80 mBq/m³ and from 1.45 to 15.1 mBq/m³ with a mean of 8.47 mBq/m³, respectively. Activity concentrations of both radionuclides at Kume-jima and Minamidaito-jima were almost the same as at Okinawa-jima. It is suggested that the relatively higher activity concentrations of both radionuclides in winter observed at the present three sites are affected by the air mass from the inland area of the Asian Continent.

Key Words : atmospheric concentration, lead-210, beryllium-7, seasonal variation, Okinawa Islands

1. Introduction

Atmospheric ^{210}Pb (half-life : 22.3 y) is one of the decay products of ^{222}Rn , (generated from the earth's crust) and exists in the atmosphere adsorbed onto aerosol particles. Radon-222 (half-life : 3.8 days) is widely dispersed in the atmosphere, and its decay products, ^{210}Pb , ^{210}Bi and ^{210}Po , have been widely utilized as powerful tracers to quantify atmospheric processes.^{1),2)} Activity concentrations of ^{210}Pb in the air have been investigated and high ^{210}Pb activity concentrations have been found only in continental air masses, since ^{222}Rn sources are practically nonexistent over the oceans. Atmospheric ^{210}Pb concentrations at the earth's surface were

found to range from 0.3 to 7.8 mBq/m³ in continental East Asia, depending on the time of year and the specific locality.³⁾

Beryllium-7 (half-life : 5.3 d) is one of the cosmogenic radionuclides produced by nuclear reactions of atmospheric oxygen and nitrogen, and is produced mainly in the lower stratosphere and upper troposphere. The rate of ^7Be production varies with cosmic radiation intensity, modulated by 11-year solar cycles. Because of the variation in production rate, fluctuations in ^7Be activity concentration in surface air are the result of multiple causes.⁴⁾

This paper focuses on the seasonal variation in the air activity concentration of ^{210}Pb and ^7Be in the Okinawa Islands. The contributions of lo-

Fig. 1 Location of the observation sites in the Okinawa Islands.

cal geochemical and meteorological conditions to the atmospheric concentrations of the two nuclides are also discussed.

2. Materials and methods

Figure 1 shows the observation sites at Kume-jima (26°N , 127°E , 59.11 km^2), Okinawa-jima (University site, 26°N , 128°E , 1208.19 km^2) and Minamidaito-jima (26°N , 131°E , 30.57 km^2) in the Okinawa Islands. These three islands do not have high mountains (the highest peak is 503 m above sea level in Okinawa-jima), and thus the air masses which pass through the islands are not influenced by the topography.

Aerosol samples were collected continuously for 7-day periods at Okinawa-jima from February 2004 to December 2010, and at Kume-jima and Minamidaito-jima from May 2008 to June 2010, on quartz-fiber filters (Pallflex, Model 2500 QAT-UP) using a high-volume air sampler (Sibata Scientific Technology, Tokyo, Japan, Model HV 500 F) at an aspiration speed of $0.5\text{ m}^3\text{ min}^{-1}$. The filter was changed weekly (usually Tuesday) and immediately sent to the University of the Ryukyus laboratory to be dried in a desiccator for at least 3 days in order to de-

cay out short-lived nuclides such as ^{214}Pb , ^{214}Bi and ^{212}Pb . The filter was folded in quarters in preparation for nondestructive gamma ray measurement.

The ^{210}Pb and ^7Be activities in the aerosol samples were determined by the presence of peaks at 46.5 keV and 477 keV (γ ray) using a low background Ge detector (Canberra BE 2825 and Intertechnique EGP 2000-15 R) with a heavy shield of lead blocks (15–20 cm). The counting duration was $(8\text{--}60) \times 10^4\text{ s}$, and overall measurement errors were estimated to be approximately 7 % or less. The detector was calibrated with a standard prepared from New Brunswick Laboratory (NBL) reference material (No. 42-1, 4.04% uranium). The ^7Be activity concentrations were corrected for radioactive decay to the midpoint of the collection period.

3. Results and discussion

Figure 2 shows the monthly averaged atmospheric ^{210}Pb and ^7Be activity concentrations at Okinawa-jima over a seven-year period, and Fig. 3 shows them at Minamidaito-jima and Kume-jima (including Okinawa-jima) over a two-year period. The ^{210}Pb activity concentration ranges were $0.06\text{--}1.98\text{ mBq/m}^3$ with a

Fig. 2 Monthly variations of ^{210}Pb and ^7Be activity concentrations in aerosols at Okinawa-jima.

Fig. 3 Monthly variations in ^{210}Pb and ^7Be activity concentrations in aerosols at three sites from 2008 to 2010.

mean of 0.78 mBq/m^3 at Okinawa-jima, $0.22\text{--}1.73 \text{ mBq/m}^3$ with a mean of 0.84 mBq/m^3 at Kume-jima, and $0.13\text{--}1.60 \text{ mBq/m}^3$ with a

mean of 0.79 mBq/m^3 at Minamidaito-jima. The activity concentrations at the present three sites were low in the summer season and

remarkably high from winter to spring, thus exhibiting a "one-peak" seasonal pattern, clearly different from the "two-peak" seasonal pattern observed at Kawasaki (lat/long: 35° N, 139° E; concentration range: 0.7–1.5 mBq/m³)⁵⁾ and Tsukuba (36° N, 140° E, 0.3–0.6 mBq/m³),⁶⁾ Japan. Although the Okinawa Islands are small in area, and the activity concentration of ²²²Rn, the parent nuclide of ²¹⁰Pb, in the air is lower than that found in mainland of Japan,⁷⁾ the activity concentration of ²¹⁰Pb observed in the surface air at the present three sites is relatively higher. This "one-peak" seasonal pattern has also been reported at many continental sites including Beijing (40° N, 116° E, 0.9–4.5 mBq/m³), Chengdu (30° N, 104° E, 1.4–7.8 mBq/m³),⁸⁾ Urumqi (44° N, 88° E, 0.4–4.6 mBq/m³), Lanzhou (36° N, 104° E, 0.3–2.8 mBq/m³) and Baotou (40° N, 110° E, 0.6–2.4 mBq/m³), China⁹⁾.

For ⁷Be, the activity concentration ranges were 1.45–14.6 mBq/m³ with a mean of 7.90 mBq/m³ at Okinawa-jima, 1.71–15.1 mBq/m³ with a mean of 8.71 mBq/m³ at Kume-jima, and 2.81–14.7 mBq/m³ with a mean of 8.81 mBq/m³ at Minamidaito-jima. This seasonal pattern is similar to that of ²¹⁰Pb.

As shown in Figs. 2 and 3, both radionuclides exhibit distribution patterns similar to each other at the present three sites. The correlation coefficient of the monthly averaged concentrations of ²¹⁰Pb to ⁷Be at Okinawa-jima over 7 years is high ($r=0.83$), as it is at Kume-jima and Minamidaito-jima (for 2 years; 0.82 and 0.73, respectively).

Although the origins of both radionuclides differ from each other, a good correlation between the activities of both radionuclides in surface air was also previously reported at Tsukuba for the period from November 1987 to

May 1992,⁶⁾ at Kanazawa (37° N, 137° E) for the period from April 1983 to January 1986,¹⁰⁾ and in the Western Pacific Ocean on board cruises from the end of August to early September in 1993 to 1994.¹¹⁾

Yamamoto et al.¹²⁾ also reported that both radionuclides in precipitation at Tatsunokuchi (36° N, 136° E), Japan showed the same pattern as that of these study sites: "one-peak," high in winter and low in summer. These studies suggest that ⁷Be levels are generally higher from winter to spring when folding of the tropopause in the mid-latitudes enhances the transfer of stratospheric ⁷Be into the troposphere,^{4), 13)–15)} that ²¹⁰Pb generated from ²²²Rn in the lower stratosphere is transported into the troposphere together with atmospheric ⁷Be and subsequently exhibit a similar seasonal variation pattern,^{10), 11), 13)} and that the lower concentration of both radionuclides in summer is controlled by maritime air masses.⁶⁾ Several studies on seasonal variation patterns of atmospheric concentrations of ²¹⁰Pb and ⁷Be at observation sites other than those listed above have been reported.^{16)–19)}

The mean activity concentration of ⁷Be found in the Okinawa Islands was higher than that previously reported in mainland of Japan. Sato et al.¹¹⁾ reported that a mean ⁷Be value of 6.3 mBq/m³ was observed in the Western Pacific Ocean (on board cruises) in September 1994. This value is almost the same as was observed at Okinawa-jima during the same period. This indicates that the air masses over the Okinawa Islands from winter to spring also include ⁷Be brought from the high latitude region of the Asian Continent where high ⁷Be levels are injected into the troposphere.

Figure 4 shows the relationship between the ²¹⁰Pb activity concentration observed at Kume-

Fig. 4 Correlation of ground-level atmospheric concentrations of ^{210}Pb and ^7Be at Kume-jima and Minamidaito-jima.

Summer (May - October)

Winter (November - March)

Fig. 5 Percent of trajectory-based wind directions in summer and winter found at Okinawa-jima from March 2011 to February 2012.

jima and that at Minamidaito-jima, and the ^7Be activity concentration observed at both sites. Although there is about 500 km between the two sites, good correlations ($r=0.87$ and $r=0.83$, respectively) between the two concentrations at both sites were obtained over the sampling period. This indicates that the well-mixed continental air mass which covers Kume-jima and Minamidaito-jima projects widely into the

Western Pacific Ocean in the winter season.

A trajectory analysis was employed to calculate the paths of air parcels to be used in an assessment of source-receptor relationships of airborne chemical species. While trajectories can be calculated either forward or backward in time, in this paper, only back trajectories are discussed. Back trajectories were calculated using HYSPLIT 4 (Hybrid Single-Particle La-

grangian-Integrated Trajectory) modeling software. This software is accessible at no charge through the Air Resource Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA).²⁰⁾

Back trajectories provide the origin of an air mass located at any given place and time. Figure 5 shows the percent of trajectory-based wind directions in summer and winter found at Okinawa-jima from March 2011 to February 2012. The data show that the air masses come mainly from the East-Southeast and South-Southwest (Pacific Ocean) in the summer, and from the West-Northwest and North-Northeast (East China sea) in the winter. Multi-year observations show that this trend is consistent.

From the considerations above and the meteorological conditions of the Okinawa Islands, the lower activity concentration of ^{210}Pb found at the present three sites in the Okinawa Islands in the summer is likely due to the air mass from the Pacific Ocean having a low activity concentration of ^{210}Pb , while the higher activity concentration in winter to spring is likely due to the air mass from the Asian Continent bringing a larger amount of ^{210}Pb , resulting in a “one-peak” seasonal pattern, clearly different from the “two-peak” seasonal pattern observed in mainland of Japan.

4. Conclusion

Seasonal variations in atmospheric ^{210}Pb and ^7Be observed in the Okinawa Islands were in the range of 0.06 to 1.98 mBq/m³ and 1.45 to 15.1 mBq/m³, respectively. The activity concentrations of both radionuclides were low in the summer and remarkably high from winter to spring, showing a “one-peak” seasonal pattern. From the results of a trajectory analysis, the ^{210}Pb found in the Okinawa Islands in the

summer is affected by the air mass from the Pacific Ocean with a low ^{210}Pb concentration, while the higher ^{210}Pb values in winter are due to the air mass from the Asian Continent bringing a larger amount of ^{210}Pb . The ^7Be found in the Okinawa Islands showed a higher value than that previously reported in mainland of Japan. This indicates that ^7Be is also delivered together with ^{210}Pb by air masses from the Asian Continent.

Acknowledgements

The authors are indebted to Mr. K. Higashi at Minamidaito-jima and Mr. S. Hukushima at Kume-jima for collecting the aerosol samples.

References

- 1) Tokieda, T., Yamanaka, K., Harada, K. and Tsunogai, S., Seasonal variations of residence time and upper atmospheric contribution of aerosols studied with Pb-210, Bi-210, Po-210 and Be-7, *Tellus*, **48 B**, 690-702 (1996)
- 2) Porstendörfer, J., Properties and behaviour of radon and thoron and their decay products in the air, *J. Aerosol Sci.*, **25**, 219-263 (1994)
- 3) Doi, T., Sato, S. and Sato, J., Atmospheric concentration of ^{210}Pb in East Asia and its contribution to Japanese Islands by long-range transport, *RADIOISOTOPES*, **56**, 115-130 (2007)
- 4) Feely, H. W., Larsen, R. J. and Sanderson, C. G., Factors that cause seasonal variations in beryllium-7 concentrations in surface air, *J. Environ. Radioact.*, **9**, 223-249 (1989)
- 5) Koike, Y., Kukita, K., Nakamura, T. and Sato, J., Atmospheric concentration of ^{212}Pb at Kawasaki, Japan, *RADIOISOTOPES*, **54**, 87-94 (2005)
- 6) Sato, J., Doi, T., Segawa, T. and Sugawara, S., Seasonal variation at Tsukuba, Japan, from the 1991 of atmospheric concentrations of ^{210}Pb and ^7Be with a possible observation of ^{210}Pb originating eruption of Pinatubo volcano, Philippines, *Geochem. J.*, **28**, 123-129 (1994)
- 7) Abe, M. and Abe, S., A nation-wide survey of ra-

- don indoors and outdoors in Japan, Radon in the living environment-levels and risks, Proceedings of the 15 th NIRS seminar, Chiba, Japan, Dec. 3-4, 79-88 (1987) (in Japanese)
- 8) Sato, S., Doi, T. and Sato, J., Atmospheric concentration of ^{210}Pb at Beijing and Chengdu, the People's Republic of China, *RADIOISOTOPES*, **49**, 439-446 (2000)
 - 9) Doi, T. and Sato, J., Variation of atmospheric ^{210}Pb concentration in the inland area of Chinese continent, *RADIOISOTOPES*, **44**, 701-709 (1995) (in Japanese)
 - 10) Tada, T., Zhao, L., Komura, K. and Sakanoue, M., Study on variations of radionuclide concentrations in atmosphere, *Chikyu Kagaku (Geochemistry)*, **20**, 98-102 (1986) (in Japanese)
 - 11) Sato, S., Murai, H., Doi, T. and Sato, J., Atmospheric concentration of ^{210}Pb and ^7Be over the Western Pacific Ocean, *RADIOISOTOPES*, **47**, 546-554 (1998)
 - 12) Yamamoto, M., Sakaguchi, A., Sakai, K., Hirose, K., Igarashi, Y. and Kim, C. K., Seasonal and spatial variation of atmospheric ^{210}Pb and ^7Be deposition, features of the Japan Sea side of Japan, *J. Environ. Radioact.*, **86**, 110-131 (2006)
 - 13) Todd, J. F., Wong, G. T. F., Olsen, C. R. and Larsen, L. L., Atmospheric depositional characteristics of beryllium-7 and lead-210 along the southeastern Virginia coast, *J. Geophys. Res.*, **94**, 11106-11116 (1989)
 - 14) Hicks, B. B. and Goodman, H. S., Seasonal and latitudinal variations of atmospheric radioactivity along Australia's east coast (150°E longitude), *Tellus*, **29**, 128-188 (1977)
 - 15) Narazaki, Y., Fujitaka, K., Igarashi, S., Ishikawa, Y. and Fujinami, N., Seasonal variation of ^7Be deposition in Japan, *J. Radioanal. Nucl. Chem.*, **256**, 489-496 (2003)
 - 16) Paatero, J., Hatakka, J. and Viisanen, Y., Trajectory analysis of ^{210}Pb and ^7Be in ground-level air in Southern Finland, *Radiochemistry*, **43**, 475-481 (2001)
 - 17) McNeary, D. and Baskaran, M., Depositional characteristics of ^7Be and ^{210}Pb in southeastern Michigan, *J. Geophys. Res.*, **108** (D 7), 4210 (2003)
 - 18) Bourcier, L., Masson, O., Laj, P., Pichon, J. M., Paulat, P., Freney, E. and Sellegri, K., Comparative trends and seasonal variation of ^7Be , ^{210}Pb and ^{137}Cs at two altitude sites in the central part of France, *J. Environ. Radioact.*, **102**, 294-301 (2011)
 - 19) Daish, S. R., Dale, A. A., Dale, C. J., May, R. and Rowe, J. E., The temporal variations of ^7Be , ^{210}Pb and ^{210}Po in air in England, *J. Environ. Radioact.*, **84**, 457-467 (2005)
 - 20) Rolph, G. D., Real-Time Environmental Applications and Display System (READY) Website (<http://ready.arl.noaa.gov>), NOAA Air Resources Laboratory, Silver Spring, MD (2010)