



Temporal Variations in Cosmogenic and Terrestrial Radionuclides

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Abstract: Two types of radionuclides, cosmogenic Be-7 (half life 53.3 days) and terrestrial Ra-226 (half life 1600 years), were measured in Tokyo, Japan in 2002-2006. Be-7 is produced by galactic cosmic rays in the upper atmosphere and Ra-226 is a decay daughter of the uranium-series radionuclide. These radionuclides in surface air were collected on a filter with a high-volume air sampler and their concentrations were determined from a gamma-ray spectroscopic experiment. We analyzed their temporal variations in measured concentrations in surface air. The Be-7 concentrations indicated seasonal variations with enhancements in spring and autumn. On the other hand, the Ra-226 concentrations show no temporal variation and almost constant. Be-7 are produced in the upper atmosphere and transported to the surface by the atmospheric motions. We suggested the air mass mixing or circulation between the lower stratosphere and upper troposphere in spring and autumn to explain the measured variations in Be-7 concentration. Ra-226 is of terrestrial origin and distributed in the atmosphere near the surface where seasonally varying atmospheric motions do not take place. It results in no temporal variations in Ra-226 concentration.

Introduction

There are two types of radionuclides of cosmogenic and terrestrial origin in the Earth's atmosphere. Cosmogenic radionuclides result from nuclear interactions of galactic cosmic rays with atmospheric nuclei and are produced in the upper atmosphere. Be-7 and C-14 are known as typical ones. On the other hand, terrestrial radionuclides are daughter radioactive nuclei of the uranium-series, thorium-series and actinium-series radionuclides. These terrestrial ones are emitted from the Earth's crust into the atmosphere and distribute in the lower atmosphere near the surface.

The cosmogenic Be-7 concentration provides information on a relation of the solar activity with galactic cosmic ray flux near the Earth and its temporal variations give a clue on air mass motions as an atmospheric tracer. Previous Be-7 experiments were carried out at several stations in the world and these results were reported [1], [2], [3], [4], [5]. In particular, temporal variations in Be-7 concentration in surface air deserve our attention. The time scale of the variations depends

on the location and ranges from a day to a month. It indicates that the air mass motions in the upper atmosphere vary with the location. On the other hand, the temporal variations in surface concentrations of terrestrial radionuclides are thought to be much less than that of the cosmogenic radionuclide because their distributions concentrate in the lower altitudes near the surface where temporal variations in atmospheric motions are very small.

We started the measurement of the cosmogenic Be-7 and terrestrial Ra-226 in surface air in Tokyo in 2002. Ra-226 (half life 1600 years) is a daughter nuclide of uranium series element of Th-230. In this paper we compare the temporal variations in the surface Be-7 with those in the terrestrial Ra-226 radionuclide and discuss the differences from a viewpoint of the air mass motions.

Measurement

We started the measurement of the cosmogenic Be-7 and terrestrial Ra-226 in surface air in Tokyo in 2002. Tokyo is located in the Mid-Asian

Pacific region where is meteorologically important but Be-7 experiments were not little reported in the past.

Cosmogenic Be-7 and terrestrial Ra-226 radionuclides are attached to small aerosol particles and collected on a filter with a high-volume air sampler. Since Be-7 (half life 53.3 days) and Ra-226 (half life 1600 years) emit gamma-ray lines at 477.53 keV and 185.90 keV, respectively, their concentrations (mBq/m³) were determined from the gamma-ray spectroscopic experiments. In the present experiment the Ge gamma-ray spectrometer with high energy resolution was used. The experimental is described in detail [6].

An example of gamma-ray spectrum in 100-600 keV is shown in Figure 1. A few gamma-ray lines were detected and their lines are identified. The Be-7 line is the strongest but the Ra-226 line is weak, as shown in Figure 1. We plotted temporal variations in surface Be-7 and Ra-226 concentrations in 2002-2006 in Figure 2 and 3, respectively. The Be-7 concentrations indicated seasonal variations with enhancements in spring and autumn. On the other hand, we do not see the significant variations in Ra-226 concentration within the experimental errors. There are different temporal variations in surface concentrations between Be-7 and Ra-226.

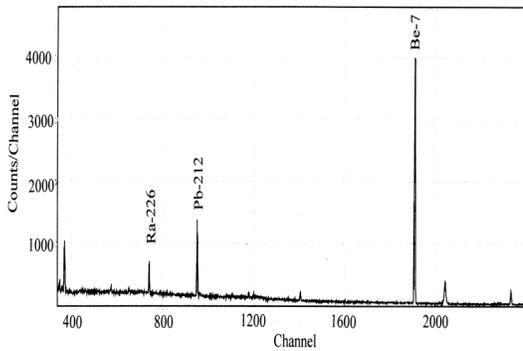


Figure 1: Example of gamma-ray spectrum in 100-600 keV.

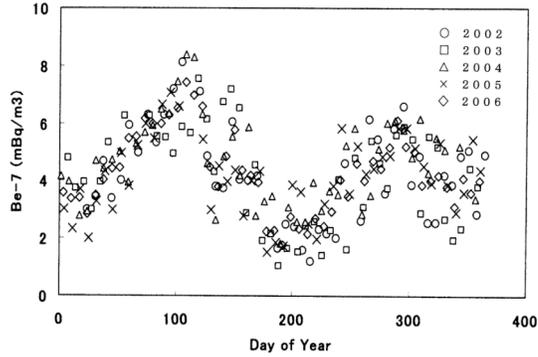


Figure 2: Temporal variations in Be-7 concentration in 2002-2006.

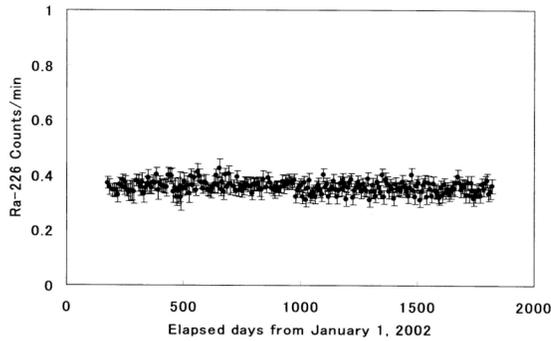


Figure 3: Temporal variations in Ra-226 concentration in 2002-2006.

Discussion

We discuss the differences in temporal variations in the surface concentrations between Be-7 and Ra-226. The Be-7 concentrations enhance in spring and autumn every year, while the Ra-226 concentrations are constant within the experimental errors. The variations in Be-7 concentrations are not caused by those in galactic cosmic ray flux and rapid increases in solar energetic particles associated with large solar flares and coronal mass ejections. The variations in surface Be-7 concentrations are due to the atmospheric effects that are closely connected with a change of meteorological conditions in Japan.

There is a difference in altitude distributions between cosmogenic Be-7 and terrestrial Ra-226 because of their different production processes. The Be-7 production rate is calculated as a function of the atmospheric depth [7], [8]. The Be-7 production rate peaks around 100-150g/cm², as shown in Figure 4. It indicates that most of Be-7 is produced in the lower stratosphere. It has been thought that there is not the efficient atmospheric circulation or mixing between the stratosphere and troposphere. However, a possibility of the stratosphere-troposphere air exchange was recently discussed [9], [10]. If this air exchange takes place, Be-7 produced in the lower stratosphere could transport to the troposphere and explain the results of high Be-7 concentrations in surface air.

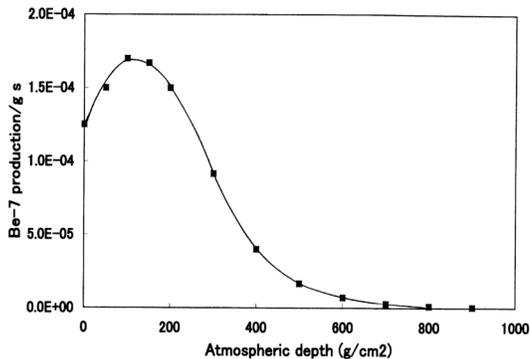


Figure 4: The Be-7 production rate as a function of the atmospheric depth.

Here we discuss a possibility of the atmospheric exchange between the stratosphere and troposphere in Japanese spring and autumn [5]. It has been well known that a westerly carries a pair of a traveling high pressure and extratropical low pressure in spring and autumn. The high and low pressures pass through Japan in a period of a few days. If there is a pair of the high and low pressures, cold air flows downwardly from the jet-stream to the surface high pressure, while the warm air flows upwardly from the surface low pressure to the jet-stream. These air mass motions cause the atmospheric circulation or mixing between the stratosphere and troposphere. It results in the enhancements of the surface Be-7 concentration [11].

On the other hand, terrestrial radionuclides are emitted from the Earth's crust into the atmosphere, suggesting that most of them distribute in the lower atmosphere near the surface. A direct measurement of the altitude distribution of Ra-226 has not been reported in the past. In stead of Ra-226, the altitude distribution of terrestrial Pb-210 that is a radionuclide of uranium series and its half life is 22.3 years was measured [12]. The measured distribution is shown in Figure 5, indicating that the Pb-210 concentration is very high near the surface. The distribution of Ra-226 also could be a similar to Pb-210 because both radionuclides are of terrestrial origin. We consider that the altitude distributions are quite different between Be-7 and Pb-210. Ra-226 near the surface should not be affected by the seasonal atmospheric circulation at the boundary region between the stratosphere and troposphere, suggesting that the Ra-226 surface concentrations are nearly constant and do not vary seasonally.

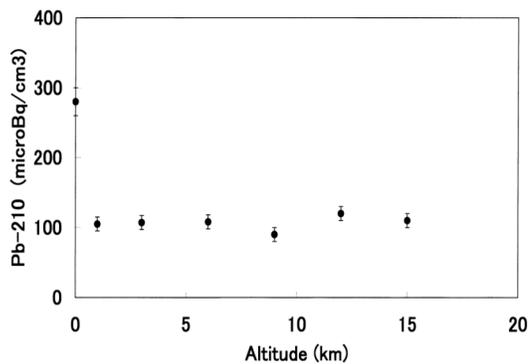


Figure 5: The altitude distribution of Pb-210 concentration [10].

As another example of the terrestrial radionuclide, we show the temporal variations in K-40 concentration in surface air in Figure 6. The K-40 is a typical terrestrial radionuclide and emits a gamma-ray line at 1460.8 keV with a half life 1.28×10^9 years. The K-40 surface concentrations exhibited were almost constant in 2002-2006. The measured surface concentrations of both terrestrial Ra-226 and K-40 do not indicate temporal

variations and are constant within the experimental errors. This result seems to support that the altitude distribution of K-40 is similar to that of Ra-226.

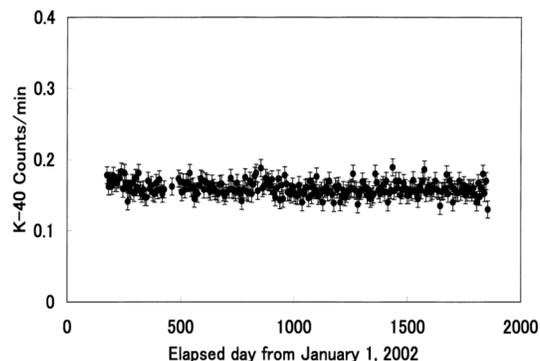


Figure 6: Variations in terrestrial K-40 concentrations in 2002-2006.

In summary, the present measurement of Be-7 and Ra-226 surface concentrations shows quite different temporal variations. This difference is considered to be due to their altitude distributions. The enhancements of Be-7 concentrations in spring and autumn are expected to be caused as follows: Be-7 in the lower stratosphere is affected by the stratosphere-troposphere exchange and brought into the troposphere, resulting in the increase of surface concentrations. On the other hand, Ra-226 emitted from the Earth's crust is concentrated near the surface and the concentrations do not vary with season.

References

- [1] J.E. Dibb. Atmospheric Deposition of Be-7 in the Chesapeake Bay Region. *J. Geophys. Res.* 94, 2261 (1989).
- [2] M. Baskaran, C.H. Coleman and P.H. Santschi. Atmospheric deposition Fluxes of Be-7 and Pb-210 at Galveston and College Station, Texas. *J. Geophys. Res.* 98, 20555 (1993).
- [3] K. Megumi, T. Matsunami, N. Ito, S. Kiyoda, A. Mizohara and T. Asano. Factors, Especially Sunspot Number, Causing Variations in Surface Air Concentrations and Depositions of Be-7 in Osaka, Japan. *Geophys. Res. Lett.* 27, 361 (2000).
- [4] J.E. Dibb, L.D. Meeker, R.C. Finkel, J.R. Southon, M.W. Caffee and L.A. Barrie. Estimation of Stratospheric Input to the Arctic Troposphere: Be-7 and Be-10 in Aerosol at Alert, Canada. *J. Geophys. Res.* 99, 12855 (1994).
- [5] M. Yoshimori. Be-7 Radionuclide as a Tracer of Vertical Air Mass Transport in the Troposphere. in the Upper Atmosphere. *Adv. Space Res.* 36, 828 (2005).
- [6] M. Yoshimori, H. Hirayama, S. Mori, K. Sasaki and H. Sakurai. Be-7 Nuclei Produced by Galactic Cosmic Rays and Solar Energetic Particles in the Earth's Atmosphere. *Adv. Space Res.* 32, 2691 (2003).
- [7] J. Masarik and J. Beer. Simulation of Particle Fluxes and Cosmogenic Nuclide Production in the Earth's Atmosphere. *J. Geophys. Res.* 104, 12099 (1999).
- [8] M. Yoshimori. Production and Behavior of Be-7 Radionuclide in the Upper Atmosphere. *Adv. Space Res.* 36, 922 (2005).
- [9] C.E. Jordan, D.E. Dibb and R.C. Finkel. Be-10/Be-7 Tracer of Atmospheric Transport and Stratosphere-Troposphere Exchange. *J. Geophys. Res.* 108, 4234 (2003).
- [10] C. Land and J. Feichter. Stratosphere-Troposphere Exchange. *J. Geophys. Res.* 108, 8523 (2003).
- [11] M. Yoshimori. Atmospheric Air Mass Motions Studied from Measurement of Cosmogenic Be-7 Radionuclide. 20th International Cosmic Ray Conference 2, 361 (2005).
- [12] L. Kownacka. Vertical Distributions of Beryllium 7 and Lead 210 in the Troposphere and Lower Stratospheric Air. *Nukleonika* 47, 79 (2002).