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Temporal Variations in Surface Concentrations of Radionuclides as Viewed from Altitude Distributions

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We measured temporal variations in surface concentrations of two terrestrial radionuclides of ^{210}Pb (half life 22.3 years) and ^{226}Ra (half life 1600 years) in Tokyo. ^{210}Pb is a decay daughter of ^{222}Rn (half life 3.8 days) and emits a gamma-ray line at 46.50 keV and ^{226}Ra is a parent nucleus of ^{222}Rn and emits a gamma-ray line at 185.90 keV. The present results indicate that the surface concentration of ^{210}Pb is seasonally variable with enhancements in spring and autumn, while ^{222}Rn exhibits a constant surface concentration. We discuss the difference in the temporal variations between ^{210}Pb and ^{226}Ra from a viewpoint on their altitude distributions.

1. Introduction

There are numerous terrestrial radioactive nuclides on the Earth's surface. For example, uranium-series nuclides (^{210}Pb, ^{214}Pb, ^{214}Bi, ^{222}Rn and ^{226}Ra), thorium-series ones (^{208}Tl, ^{212}Pb and ^{212}Bi), actinium-series one (^{235}U) and ^{40}K are well known. These terrestrial radioactive nuclides are emitted from the Earths crust into the atmosphere and are expected to be distributed in the lower troposphere. The surface concentration of ^{210}Pb were measured along with ^{7}Be [1], [2], [3], [4], [5], and the atmospheric transport process and the residence time were simulated [6], [7], [8], [9]. Although a correlation between the surface concentrations of ^{7}Be and ^{210}Pb were studied, the surface concentration of ^{210}Pb depends on location and season. The concentrations of ^{210}Pb from the continents were reported to be much higher than that from the oceans because the ^{222}Rn is a continent source. On the other hand, the surface concentration and altitude distribution of ^{222}Rn was not enough reported.

In this paper we describe the measurements of the surface concentrations of two terrestrial radionuclides of ^{210}Pb and ^{226}Ra. These two radionuclides are both uranium-series ones and related with the decay processes of ^{226}Ra → ^{222}Rn → ^{218}Po→ ^{214}Bi (^{214}Pb) → ^{210}Pb. The experiment started in Tokyo (35N/139E) in January 2002 for ^{210}Pb and in June 2003 for ^{222}Rn, and their temporal variations were studied. We discuss their altitude distributions from the measured temporal variations.

2. Measurement

We have continuously measured the surface concentrations of ^{210}Pb and ^{226}Ra using a high-volume air sampler. The air-flow rate of the air sampler is 1 m^3 per min and natural radionuclide-borne aerosol particles are collected on a glass fiber filter (25 cm x 20 cm). The radionuclides are sampled for one week (totally collected air volume is 10^4 m^3 per week) and a gamma-ray lines emitted from radionuclides are measured with a high-resolution Ge spectrometer. The measurement and method are given in detail [5].

A typical gamma-ray spectrum of the collected radionuclides is shown in Figure 1. Numerous gamma-ray lines of cosmogenic and terrestrial radionuclides are detected. Although the intensities of gamma-ray lines of ^{210}Pb (46.50 keV) at 180 channel and ^{226}Ra (185.90 keV) at 740 channel are low, these lines are clearly discriminated from the background. The expanded spectrum of the ^{210}Pb and ^{226}Ra line region below 1000 channel (250 keV) is shown in Figure 2.

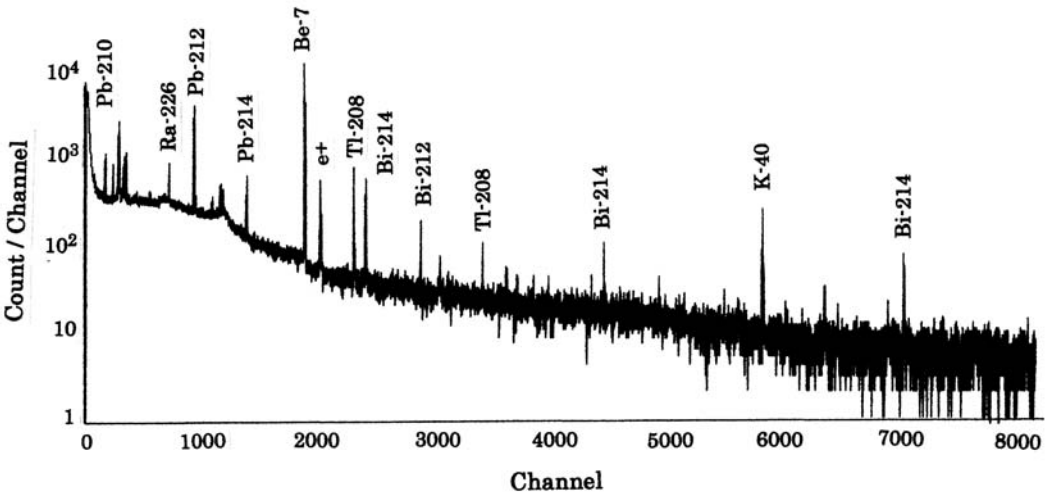

Figure 1. A typical gamma-ray spectrum of the collected radionuclides.

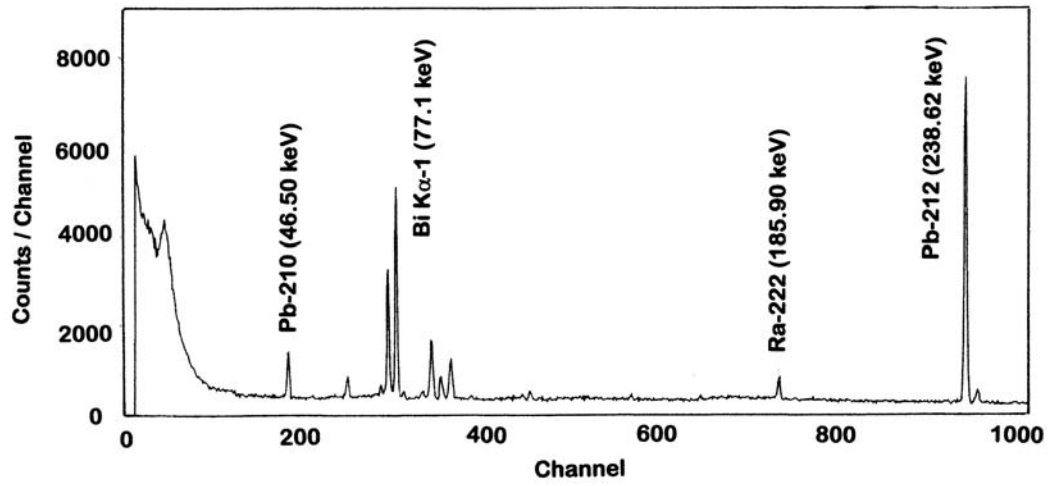

Figure 2. ^{210}Pb (46.50 keV) and ^{226}Ra (185.90 keV) lines.

The temporal variations in the count rate of ^{210}Pb line measured in January 2002 to June 2005 are plotted in Figure 3. The count rates vary with time and exhibit the enhancements in spring and autumn every year. The count rate of 1 count/min corresponds to the ^{210}Pb radioactivity of 0.83 mBq/m^3. The temporal variations in the ^{210}Pb count rate is similar to those in the ^{7}Be [10]. The temporal variations in the count rate of ^{226}Ra line in June 2003 to June 2005 are shown in Figure 4. The count rate is almost constant without seasonal variations and the temporal variations are quite different from those of ^{210}Pb. The count rate of 1 count/min corresponds to the ^{226}Ra radioactivity of 0.75 mBq/m^3.

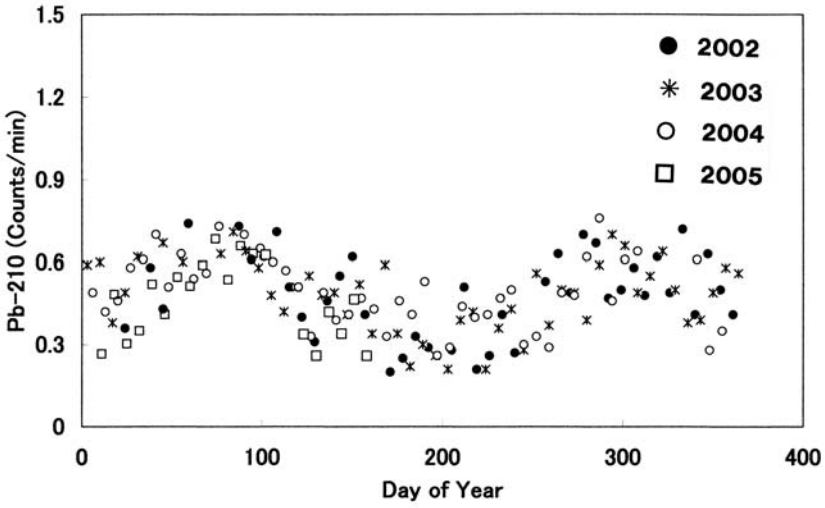

Figure 3. Temporal variations in count rate of ^{210}Pb line at 46.50 keV.

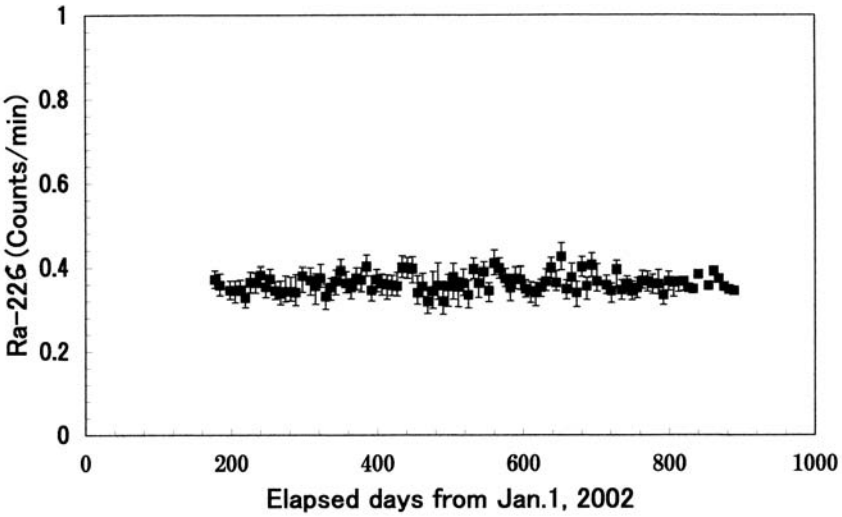

Figure 4. Temporal variations in count rate of ^{226}Ra line at 185.90 keV.

3. Discussion

We discuss the difference in temporal variations between ^{210}Pb and ^{226}Ra from a view point of the altitude distribution. Air mass motions that affect the surface concentration vary with the altitude. If the terrestrial radionuclides have similar altitude distributions, their temporal variations in the surface concentrations are expected to be similar. In general, the terrestrial radionuclides are thought to be distributed in the lower troposphere. It indicates that their surface concentrations could not vary with time. However, the present measurement shows that the surface count rate of ^{210}Pb is enhanced in spring and autumn though that of

^{226}Ra is constant with time. ^{210}Pb exhibits similar temporal variations to ^{7}Be that are mostly produced at high altitudes (lower stratosphere and higher troposphere). The present result may suggest that the distribution of ^{210}Pb expands to higher altitudes.

Here we consider the possibility that ^{210}Pb moves up to higher altitudes. ^{210}Pb is the decay daughter of ^{222}Rn. The Earth's crust contains the long-lived ^{238}U that decays over ^{226}Ra to ^{222}Rn. ^{222}Rn emanates out of the Earth's crust into the atmosphere. Since ^{222}Rn is a water-insolvent noble gas, part of ^{222}Rn could move up to the upper atmosphere without attachment to an aerosol particle and decay into ^{210}Pb. Since ^{210}Pb is metallic, it is attached to a small particle at high altitudes and falls to the surface without decay (half life of 22.3 years). There is the measurement that supports the high ^{210}Pb concentration at high altitudes. It indicated that the ^{210}Pb concentration does not decrease with altitude but is nearly constant (about one third of the surface concentration) above 1 km [10]. On the other hand, metallic ^{226}Ra in the Earth's crust escapes directly into the atmosphere and is attached to the aerosol particle. Therefore, the distribution of ^{226}Ra should be restricted in the lower troposphere.

The enhancement of surface ^{7}Be concentration in spring and autumn is thought to be caused by the air mass mixing between the stratosphere and troposphere or the efficient vertical transport in the troposphere [11], [12], [13]. If we assume such atmospheric motions, the measured enhancement of the surface count rate of ^{210}Pb in spring and autumn could be explained from the assumption of distribution of ^{210}Pb at high altitudes.

4. Acknowledgements

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