Variation of Atmospheric ^{7}Be and ^{210}Pb Depositions at Fukuoka, Japan

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INTRODUCTION

Beryllium-7 (half-life 53.29 d) is one of the radionuclide produced by spallation reactions of cosmic rays with light atmospheric nuclei, such as carbon, nitrogen and oxygen. Approximately 70% of ^{7}Be produced in the stratosphere, with the remaining 30% produced in the troposphere. A residence time is estimated about a year in the stratosphere, and about six weeks in the troposphere. Most of the ^{7}Be that are produced in the stratosphere don't reach the troposphere except during spring when seasonal thinning of the tropopause takes place at midlatitudes, resulting in air exchange between stratosphere and troposphere. ^{7}Be rapidly associates primarily with submicron-sized aerosol particles. Gravitational settling and precipitation processes largely accomplish transfer to the earth's surface. ^{7}Be associated with aerosol particles is an ideal tool with which to study atmospheric transport processes.

Lead-210 (half-life 22.3 y) which is one of the natural radionuclide of the ^{238}U decay series is widely used as a tracer. ^{222}Rn (half-life 3.8 d), noble gas of the ^{238}U decay series, easily diffuses out mainly from land surface to the atmosphere via a series of short-lives secondary nuclides and becomes irreversibly attached to submicron-sized aerosols. ^{210}Pb depositional pattern gave us information on continental aerosols in lower troposphere.

These two radionuclides with their different sources are therefor useful to understand the mechanisms of aerosol removal from the atmosphere. The activity of these radionuclides in the precipitation is expected to vary with location and time. These radionuclides have measured routinely in many places of the world in order to study the description of environmental processes such as aerosol transit and residence times in the troposphere, aerosol deposition velocities and aerosol trapping by ground vegetation (3,10). The data for most of sampling sites show seasonal variations in the concentration of ^{7}Be. The relationship between precipitation and deposition of ^{7}Be has often been studied (1,2,4,5,6,7,9). Seasonal variations in the concentration of ^{7}Be in surface air have often been attributed to the influence of variations in the rate of exchange of air between the stratosphere and the troposphere. Other factors influenced to seasonal variation are the rate of vertical mixing within the troposphere, the rate of poleward transport of air masses from middle latitudes to high latitudes and the rainfall rate indicating the importance of washout of the atmospheric aerosol.

In order to estimate the deposition pattern of ^{7}Be, the model has been considered and was applied to the data presented in this paper. The Kyushu Island is located at the West End of Japan and also at the East End of Asian Continent. This site is important for the study of the effect with the continental aerosol transport.

EXPERIMENTAL AND METHOD

Samples have been collected at Fukuoka in Japan. Deposition samples including rain and atmospheric deposition (wet deposition and dry deposition) were collected routinely throughout every month with a collector which has a surface area of 0.5 August 1994 to October 1999. The collected deposition samples were evaporated to dryness. Activity of ^{7}Be and ^{210}Pb in each sample was measured by non-destructive gamma-ray spectrometry using gamma rays of 478 keV and 46.5 keV, respectively. Aerosol samples have been collected using high volume air sampler with pumping rate of 1000 l/min to estimate wet and dry deposition rate.

RESULTS AND DISCUSSION

The results for monthly ^{7}Be and ^{210}Pb depositions and precipitation are shown in Fig. 1. The depositions of ^{7}Be and ^{210}Pb during these periods vary by more than a factor of 10 and 6, between 1.11 and 11.54, and 0.19 and 1.31 Bq/m^{2}/d, respectively. There were seasonal variations of ^{7}Be and ^{210}Pb depositions: high in spring (from March to May) and in winter (1996 and 1997), while monthly precipitation were high in summer. Recently, the variance became larger in both depositions and precipitation. Seasonal variation of each radionuclide is the same, however, is not the same from year to year.

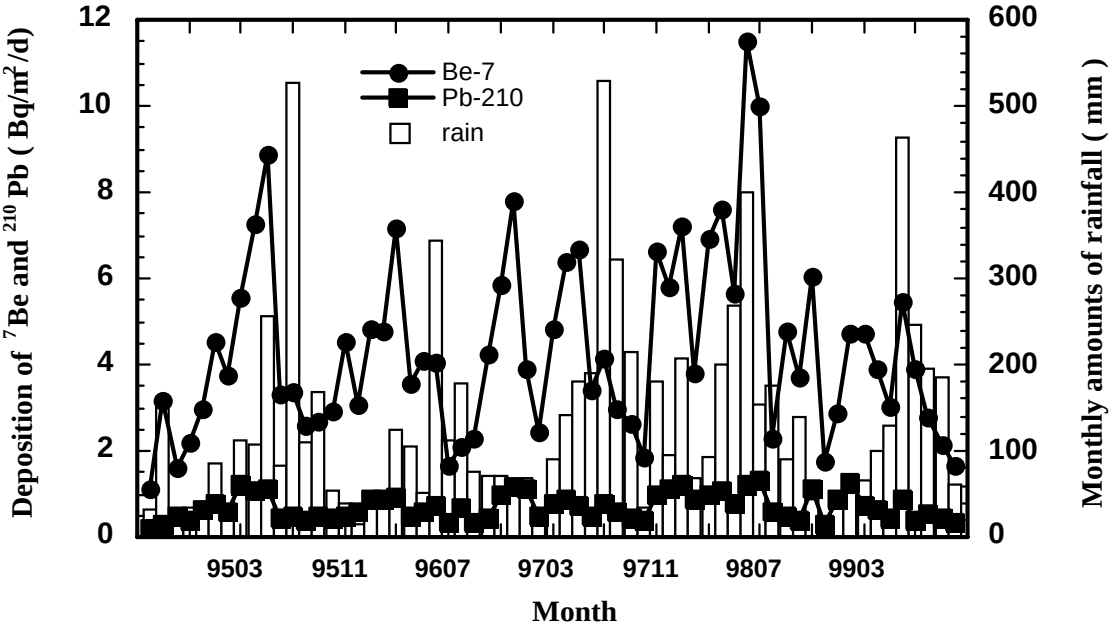


Fig. 1. Depositions of ^{7}Be and ^{210}Pb and montly amounts of rainfall at Fukuoka.

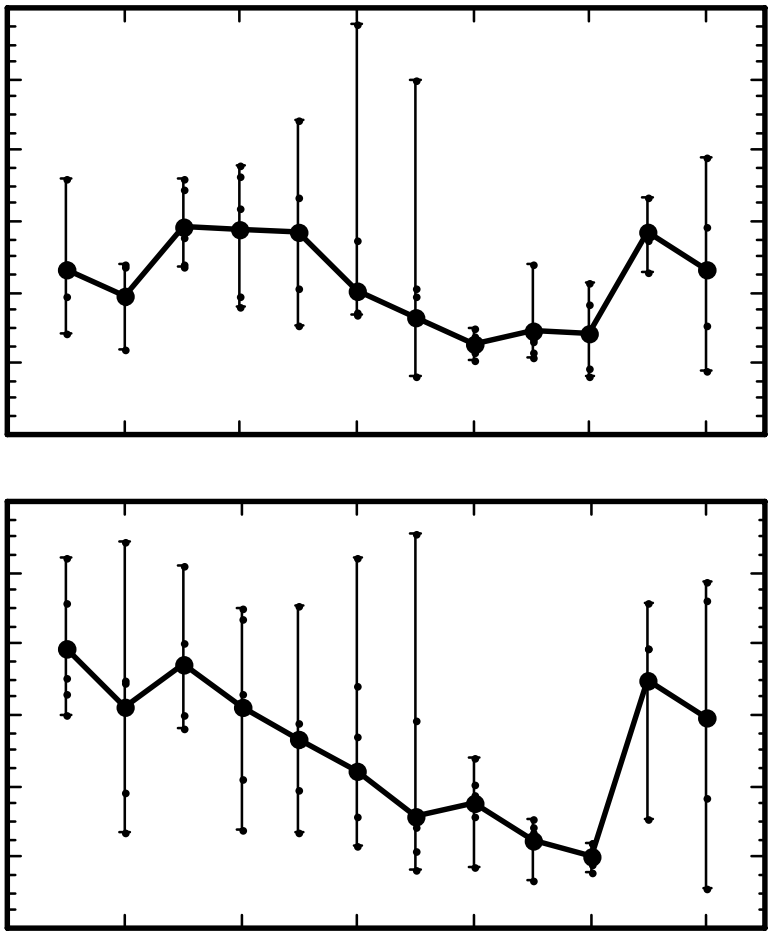
Average depositions in each calendar year are listed in Table 1. The total amounts of rainfall in 1997 and 1998 were larger than that in 1995 and 1996. Larsen have observed that the concentrations of ^{7}Be in surface air samples have been decreasing under the influence of the solar activity (8). It has been known that the galactic cosmic-ray intensity varies with solar activity. The 11-year solar cycle of cosmic ray plays an important role of long-term variation on the yearly average concentration of ^{7}Be. There is however no tendency to decrease or increase in annual depositions of ^{7}Be in this period. One explanation for this result may be that the deposition pattern of ^{7}Be was more affected by the variation on meteorological condition than the solar cycle of cosmic ray.

Table 1. Average ^{7}Be and ^{210}Pb deposition at Fukuoka

year	^{7}Be (Bq/m^{2}/d)	^{210}Pb (Bq/m^{2}/d)	annual rainfall (mm)
1995	4.38	0.67	1593
1996	4.38	0.71	1275.5
1997	4.31	0.73	2083
1998	5.95	0.88	1865.5

The monthly average depositions of ^{7}Be and ^{210}Pb for 5 years are shown in Fig. 2. Winter peak (from November to March) and minimal in fall (from August to October) of ^{7}Be and ^{210}Pb and spring peak (from April to May) of ^{7}Be were observed. Spring peak of ^{7}Be is a well-known phenomenon resulting from the increase in the rate of transport of stratospheric air into troposphere. It is considered that the reason of winter peak was the effect of seasonal variations in the horizontal transport of tropospheric air masses from high latitudes into middle latitudes, i.e. Continental Siberian Air Mass.

^{7}Be is of cosmogenic origin and its production rate is high in the upper troposphere, ^{210}Pb, on the other hand, has a high production rate in the lower troposphere. Owing to the different sources of ^{7}Be and ^{210}Pb, the depositional pattern of both radionuclides can provide information on the specific process, which control the seasonal variation. The higher values of ^{7}Be relative to ^{210}Pb during spring season can be attributed to the increased vertical transport rate of ^{7}Be from the upper troposphere to the middle and low troposphere. Seasonal variations in the concentration of ^{7}Be in surface air depend on the rate of stratosphere- troposphere exchange of air, the rate of vertical mixing within the troposphere, and the rate of horizontal transport of air masses. The vertical mixing within the troposphere causes relatively high concentration of ^{7}Be and low concentration of ^{210}Pb in surface air. The horizontal transport of air masses also attributes to the ^{7}Be/^{210}Pb ratio. The oceanic air masses bring lower amounts of ^{210}Pb compared to the continental air masses.



The fallout composition divided into two fractions; wet and dry deposition. Wet deposition has also two scavenging mechanisms of radionuclides and aerosol in air by precipitation. In general, the scavenging mechanisms, namely, washout and rainout, are important. It is not clear that which mechanism more influences to the fallout of radionuclide. The scavenging mechanism in air may depend on the difference in precipitation mode, i.e. rain or snow, wind speed and direction, height of cloud.

The relationship between radionuclide deposition and precipitation has generally been expressed by a linear equation (6). In order to estimate the ^{7}Be deposition, we used the simple model assuming the wet deposition including the rainout and washout effects and the dry deposition were the removal process of aerosol from the atmosphere. The ^{7}Be deposition (D_{Be}) is estimated in the following model:

$$D_{Be} = A \times P + B \times R_N + C_1 \times N_F + C_2 \times N_R$$

where A, B and C are the rainout, washout and dry deposition coefficients, respectively. P is the amount of precipitation. R_N is the number of rain event, N_F is the number of rainless day and N_R is the number of rainy day. The value of these coefficients, A and B depend on the rainfall intensity. The washout and rainout coefficients were calculated by fitting of the wet deposition data. The dry deposition coefficients were derived from the dry deposition measurements. The results of this model application to the monthly ^{7}Be depositions are depicted in Fig. 3. The model calculated values are in agreement with observed value. In detail observation, some differences are appeared in abnormal meteorological conditions.

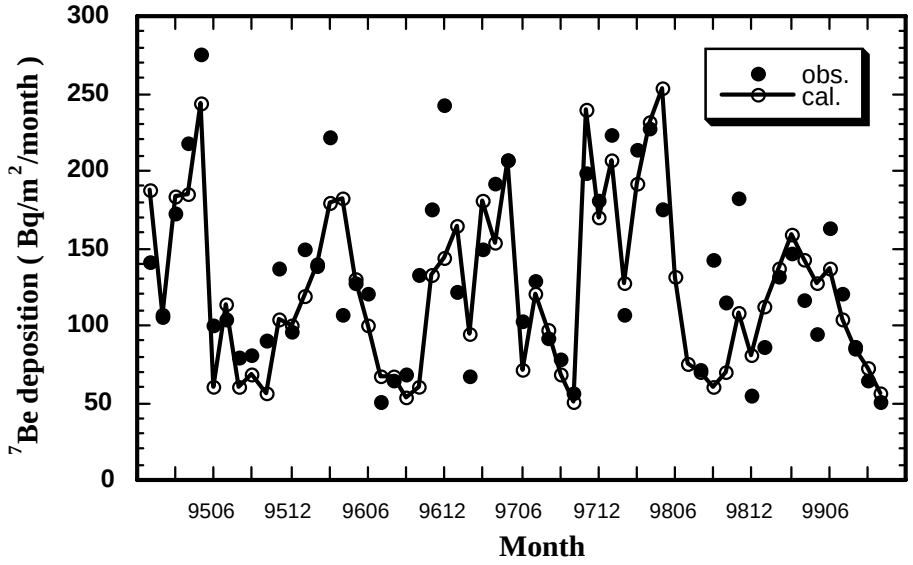


Fig. 3. Seasonal variations of model calculated and observed ^{7}Be depositions in 1995-1999.

CONCLUSION

There are seasonal variations of ^{7}Be and ^{210}Pb activity on the deposition samples in Fukuoka: high in winter (^{7}Be and ^{210}Pb) and spring (^{7}Be). The higher values of ^{7}Be and ^{210}Pb during winter (November to February) are attributed to the increased horizontal transport rate from the continental region of higher latitudes. There is an increased transfer of ^{7}Be from the stratosphere to the troposphere in spring. The good correlation was observed between ^{7}Be and ^{210}Pb activities and the amount of precipitation. But this correlation depends on the season. These two radionuclides show the same mechanism of aerosol removal from the atmosphere in spite of their different sources. The fallout composition divided into two fractions: wet deposition and dry deposition. The wet deposition was major process to remove the aerosol from the atmosphere. The scavenging by the washout mainly contribute to the removal process. The relationship of ^{7}Be and ^{210}Pb deposition pattern is almost similar, indicating utility as a useful tool of atmospheric tracer. We applied the prediction model. The calculated data are in good agreement with the observed data. It needs further investigation to elucidate the relationship between the washout and rainout effects.

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