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Factors, especially sunspot number, causing variations in surface air concentrations and depositions of ^{7}Be in Osaka, Japan

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Abstracts. We found that increasing sunspot number caused a significant negative effect on monthly and yearly average air concentration and yearly deposition of ^{7}Be. For more than 15 years monthly average surface air concentration and monthly deposition of ^{7}Be was measured in Osaka, Japan. The maximum monthly average surface air concentration of ^{7}Be was 1.6 times higher than the minimum concentration through the fifteen year measurement span. Increase of several percent in the monthly averaged surface air concentration and monthly deposition of ^{7}Be were observed in the monthly mean for February-March-April 1987 period following the 23 February supernova explosion in the Large Magellanic Cloud. The seasonal variation pattern which includes the lowest surface air concentration of ^{7}Be in summer is due to the transport of air masses from low latitudes carrying low ^{7}Be concentrations into the middle latitude region.

Introduction

The importance of using naturally occurring radionuclides as environmental tracers in geophysical and geochemical studies is well known. Beryllium 7 is a relatively short-lived, cosmogenicaly-produced radioactive nuclide which is formed by spallation processes of light atmospheric nuclei. This isotope is produced, when elements such as C, N and O absorb protons and even neutrons of the primary component of cosmic rays [Lal, 1967]. ^{7}Be is recognized as an atmospheric tracer because its measurement gives information about diffusion properties of the high troposphere and low stratosphere as well as a description of environmental processes such as precipitation, washout, atmospheric particle deposition and depositional patterns of airborne contaminants [Feely et al., 1989, Matsunami and Megumi, 1994, Abe, 1995].

Since 1983, our laboratory in Osaka, Japan has monitored γ emitting radionuclides, especially ^{7}Be, in ground-level air as well as deposition. The purpose was to examine artificial radioactive contamination in the environment and to study characteristics of the environmental processes [Matsunami and Megumi, 1994]. This paper describes the effect of changes in sunspot number on 1) monthly and 2) yearly average surface air concentration and 3) the yearly deposition of ^{7}Be. Other factors causing variations in the surface air concentration and deposition of ^{7}Be such as transporting air masses, precipitation amount and super novas are also mentioned.

Measurements

Monthly average surface air concentration and monthly deposition of ^{7}Be have been measured at our laboratory in Osaka(Sakai 135.30 E, 34.32 N). Every month, airborne dust samples of aerosol particles smaller in size than 10 μ m have been continuously collected using a low volume air sampler (20 Liter/Min) with a Millipore AA filter(1.0 μ m) on the roof of our laboratory(3 M above ground). Monthly deposition samples have been collected continuously with a stainless steel basin having a 0.5 m^2 collecting area that was placed on our laboratory roof. Distilled water was poured into the basin on the first day of the month and on the last day, the collected deposition was evaporated to dryness in a polystyrene vessel. Both the dust samples and the residue samples were subjected to γ ray spectrometry. ^{7}Be and ^{137}Cs concentrations were measured using a Ge(Li) detector that was shielded with a thin brass and a 10 cm thick iron lining lead: and connected to a multi channel analyzer that measured γ rays of 477.6 keV, and γ rays of 661 keV of ^{137m}Ba, respectively [Matsunami and Megumi, 1994].

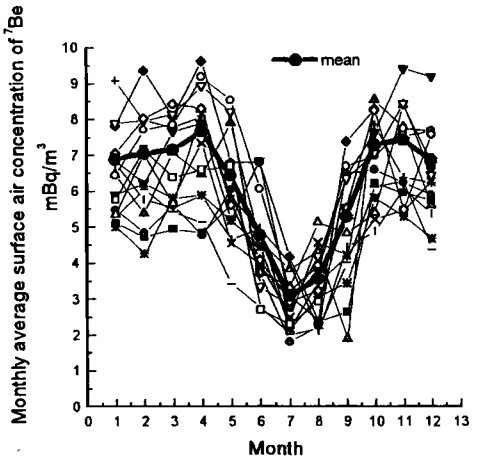

Figure 1. The seasonal variation in surface air concentration of ^{7}Be observed from 1983 to 1997 in Osaka.

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Table 1 Average wind direction and speed at atmospheric pressure 850 hPa from 1960 to 1990 observed at Shionomisaki observatory [Chronological Scientific Tables, 1997].

Month	Direction	Speed m/sec
Jan.	W	9.4
Feb.	W	8.3
Mar.	W	6.4
April	W	3.1
May	W	2.0
June	WSW	3.0
July	WSW	2.8
Aug.	S	1.5
Sept.	S	1.1
Oct.	NNW	0.7
Nov.	W	4.6
Dec.	WWN	8.6

Result and Discussion

The monthly average surface air concentrations of ^{7}Be from 1983 to 1997 are plotted in Fig.1. The seasonal variations show the lowest concentrations of ^{7}Be in summer. Table 1 lists the surface air concentration showing that, during summer, low levels correlated with the direction and mean speed of seasonal wind at the atmospheric pressure of 850 hPa observed at Shionomisaki (135.45 E, 33.26 N) meteorological observatory [Chronological Scientific Tables, 1997]. In the summer, the wind blows from the south or the west-south-west, transporting air masses from low latitudes. It is well known that the air mass from low latitudes carries low concentrations of ^{7}Be [Feely et al., 1989]. Surface air concentrations exhibited obvious spring peaks in contrast to unnoticeable autumn peaks due to influences on the exchange rate between the stratosphere and the troposphere, as described by Feely et. al. [1989] and Abe [1995].

A clear seasonal variation pattern in monthly deposition of ^{7}Be was not observed due to its dependency on the amount of the monthly precipitation, as described later. Then, the ratio of monthly deposition of ^{7}Be to the monthly precipitation amount or to the monthly precipitation frequency are listed in Table 2 , respectively. Monthly deposition of ^{7}Be was low during the summer and high during the winter. The high deposition rate during winter was the result of the relatively small amount of monthly precipitation and the subsequent large contribution of dry deposition to the total.

Table 2 Average amount and rate of ^{7}Be monthly deposition, and average monthly precipitation from 1983 to 1997

Month	Average amount of ^{7}Be monthly deposition MBq/km^{2}·month	Average monthly ^{7}Be deposition rate MBq/km^{2}.mm/ month	Average Monthly ^{7}Be Deposition rate MBq/km^{2} time	Average monthly precipitation mm/month
Jan.	79.44(41 5-127 1)	2.18(1.1-3.4)	15.3(8 9-19.8)	42 43(18-92)
Feb.	74.38(37 2-224.8)	2.04(0 6-4 7)	13.5(7.2-28.0)	53 33(9-169)
Mar.	156.24(91.1-247 5)	1 83(0.9-2.4)	18 0(8.4-28.6)	95.67(45-137)
April	122.37(52.8-172.5)	1.60(0.9-2 5)	15.5(8.6-28.4)	89.53(38-170)
May	176.65(78.4-218 5)	1.47(0.8-2 0)	21.4(9 8-31.2)	132.93(51-317)
June	157.79(68 0-232.7)	1.13(0.4-2.3)	17.0(6.8-25.4)	182.27(51-350)
July	97.72(41 9-193.6)	0.84(0.3-1.3)	11.5(4 5-22.7)	129.00 (22-225)
Aug.	68.37(16 2-175.5)	1.06(0.3-2.8)	11.7(6.0-35.9)	80.27(25-219)
Sept	113.35(56 2-194.5)	1.19(0.3-1 7)	12.7(5.1-24.3)	166 93(20-432)
Oct.	104.02(40.9-257.2)	1 48(0.8-2.5)	18.5((4.5-28.3)	88.20(21-239)
Nov.	88.85(37 9-206 5)	2.03(0.7-3.8)	15.3(9.1-26.9)	65.33(14-178)
Dec.	74.05(34 4-105 4)	2.61(1.00-5.3)	12.4(5.4-21.2)	39.87(10-73)

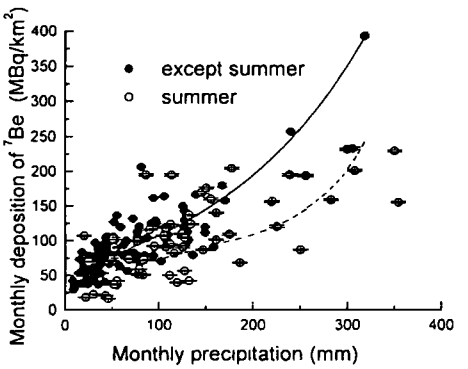

Figure 2. Correlation between monthly deposition amount of ^{7}Be and monthly precipitation amount for winter, spring and autumn, and for the summer(June, July, Aug., Sept) from 1983 to 1997 in Osaka..

As shown in Fig.2, the monthly deposition of ^{7}Be related positively with the monthly precipitation for the seasons except summer (correlation factor 0.80) and for the summer(June, July, Aug., Sept.). The maximum dry deposition of ^{7}Be may be inferred from the intercept point with zero precipitation, namely about 30 Bq/km^{2} for the summer and about 40 Bq/km^{2} for the other seasons. As Feely et al.[1989] reported, wet scavenging of ^{7}Be is the predominant process in deposition.

Figure 3 shows the relation of the monthly deposition rate of ^{7}Be for the seasons except summer, to monthly precipitation. The rate was obtained by dividing the monthly deposition amount of ^{7}Be by the monthly precipitation. One of the monthly deposition amounts was obtained by subtracting the dry deposition amount of 15 MBq/km^{2}/month. The deposition rate shows clear decreases in accordance with increases in monthly precipitation. The surface air concentration of ^{7}Be also decreased in correlation with monthly precipitation frequency(> 1mm/month) for winter and autumn. The surface air concentrations were calculated by subtracting the influence of the sunspot number described later. From the above fact, we infer that ^{7}Be wet deposition chiefly depends on wash-out, though it is necessary to directly measure the dependency of precipitation concentration of ^{7}Be on

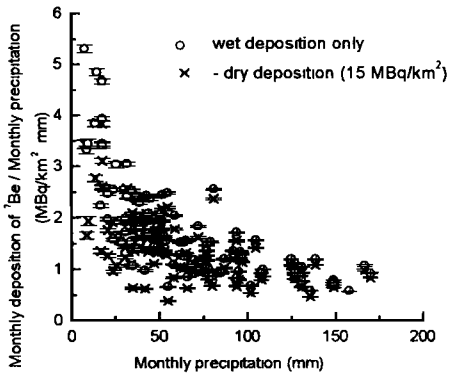

Figure 3. Correlation between monthly deposition rate of ^{7}Be for the seasons other than summer and monthly precipitation from 1983 to 1997 in Osaka.

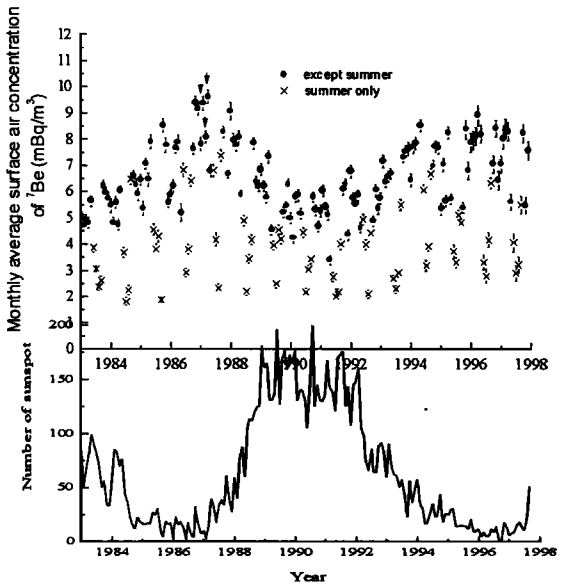

Figure 4. Variation of monthly average surface air concentration of ^{7}Be for the seasons other than summer or for the summer only and monthly sunspot number.

the precipitation rate. It is well known that wet deposition of the radon daughter nuclides chiefly depends on the effect of rain-out because the free part of ^{218}Po from aerosol particles, the daughter nuclides of ^{222}Rn, are easily co-opted by cloud particles. On the contrary, ^{7}Be normally attaches to aerosol particles in clouds and the contribution of wash-out to wet deposition of ^{7}Be may be larger than rain-out.

Figure 4 shows the monthly changes in surface air concentration of ^{7}Be in seasons other than summer and the monthly sunspot number. A clear reverse-correlation is evident between the surface air concentration of ^{7}Be and monthly sunspot number (Fig.5). This may be explained by the influence of galactic cosmic ray strength on sunspot number as

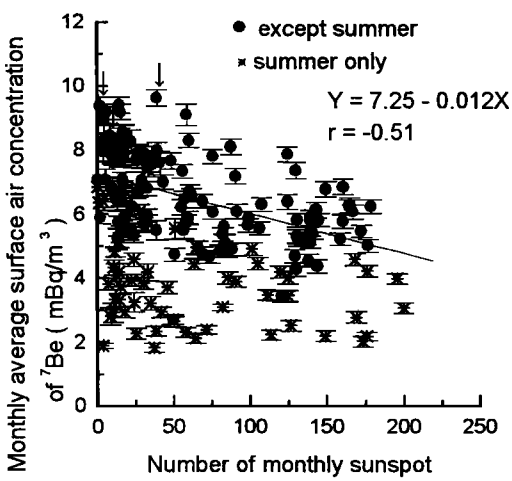

Figure 5. The relation of monthly average surface air concentration of ^{7}Be, in the season other than summer, and in summer to the monthly sunspot number.

Hotzl et al. [1991], Larsen[1993] and Matsunami and Megumi, [1994]. In Fig. 6, reverse-correlation is clearly shown between annual sunspot number and annual average air concentration of ^{7}Be or its annual deposition, respectively. However, the amount of ^{7}Be deposition in 1994 was abnormally low because of an extremely small amount of annual precipitation (~500 mm). Dependency of the yearly change in surface air concentration and monthly deposition of ^{7}Be on amount of annual precipitation is also given.

A supernova (SN) explosion occurred in the Large Magellanic Cloud on 23 February 1987. Papastefanou [1991] observed increased ^{7}Be concentration in air in Greece after the SN 1987. We observed increases in ^{7}Be

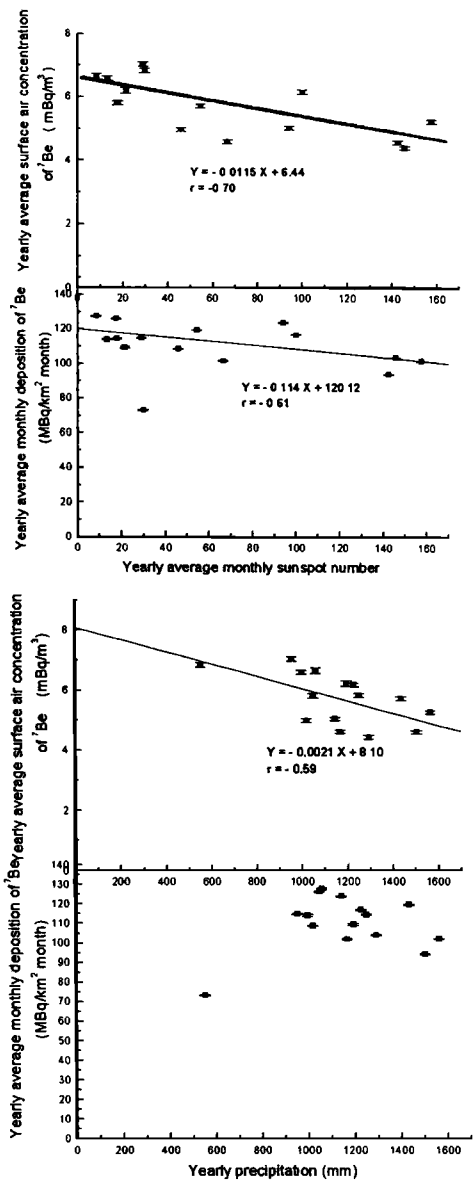

Figure 6. a). The relation of annual average surface air concentration and annual average monthly deposition of ^{7}Be to the annual average monthly sunspot number.
b). The relation of annual average surface air concentration and annual average monthly deposition of ^{7}Be to the annual precipitation.

Table 3 The mean of monthly deposition and monthly average surface air concentration of ^{7}Be and the mean of monthly precipitation and monthly sunspot number for Feb., Mar and April.

Year	Mean of monthly Deposition of ^{7}Be MBq/km^2 ·month	Mean of monthly average surface air concentration of ^{7}Be MBq/m^3	Mean of monthly Precipitation mm/month	Mean of number of monthly Sunspot
1983	111.7 ± 1.1	4.83 ± 0.20	91.3	66 1
1984	100.1 ± 1 1	5.07 ± 0.22	49.3	79 5
1985	136.7 ± 1.5	6.31 ± 0.24	123.3	16 4
1986	130.3 ± 1.2	7.26 ± 0.26	64.3	19 0
1987	146.2 ± 1,1	9.03 ± 0.25	68.7	18 9
1988	107.6 ± 1.0	7 95 ± 0.24	63.3	68 1
1989	134.8 ± 1.1	6 46 ± 0.23	113.0	142.4
1990	97.6 ± 1 0	5 32 ± 0.22	103.7	137 0
1991	108.1 ± 1 0	5.53 ± 0.21	108 0	149 8
1992	136.6 ± 1.2	5.73 ± 0 21	100 3	122 5
1993	80 6 ± 0.9	6.72 ± 0.22	54 0	74.3
1994	94.2 ± 1.0	8.25 ± 0.24	55 3	27 8
1995	122.4 ± 1.1	6.99 ± 0.25	60.7	25.0
1997	137.5 ± 1.8	8 26 ± 0.37	71 0	10 6

concentration in surface air in February, March and April of 1987 as shown with arrows in Figures 4 and 5. The surface air concentration and the deposition of ^{7}Be in this season are affected by the spring mixing especially, the sunspot number and the precipitation amount. Then, Table 3 shows clearly that the mean of monthly deposition and monthly averaged surface air concentrations of ^{7}Be for the three months of February, March and April are the highest in 1987, with reference to the mean of monthly precipitation and number of sunspot for the three months.

The 26th nuclear explosion test conducted by China in 1980 was the last atmospheric explosion test in the northern hemisphere, the 23rd, 24th and 25th atmospheric test by China were carried out in 1978[Natural Resources Defense Council, 1971-96]. These explosions led to the need for a stratospheric inventory of radioactivity. In subsequent years, the troposphere effect dissipated because of the tropospheric residence time of the radioactive aerosol is usually shorter than one month due to its efficient removal by precipitation. The ^{137}Cs deposition due to atmospheric nuclear explosion was almost complete due to stratospheric fallout, soil resuspension was negligible. Figure 7 shows the relation between the monthly depositions of ^{137}Cs and ^{7}Be. A rather good correlation was found between ^{137}Cs and ^{7}Be monthly depositions in 1979, 1981 and 1982. This may be because ^{137}Cs also was brought from the upper part of the troposphere and lower stratosphere and not by

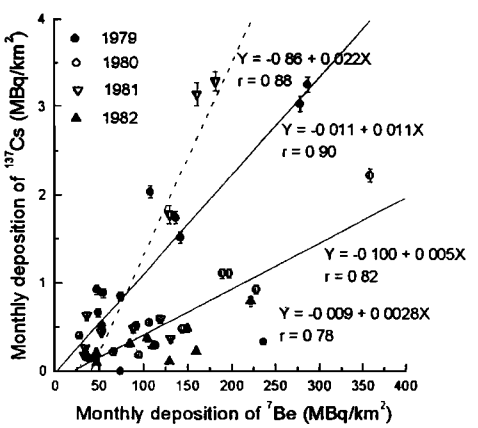

Figure 7. Correlation between the monthly deposition amount of ^{137}Cs and ^{7}Be in the following year after atmospheric nuclear explosions tests.

resuspension from the ground, which is different from the other years. We confirmed that ^{7}Be is an index for monitoring air pollution of the upper part of the troposphere and the lower part of the stratosphere.

Conclusion

Monthly average surface air concentrations and monthly deposition rate of ^{7}Be tended to be low in summer because of the transport of air masses from low latitudes in summer. Monthly average surface air concentrations of ^{7}Be in the other seasons had a negative correlation with sunspot number. The annual average surface air concentration of ^{7}Be and annual deposition of ^{7}Be also showed a negative correlation to annual sunspot number except for 1994. An increase in surface air concentrations and deposition of ^{7}Be were confirmed after a 1987 super nova.

Our results show that deposition of ^{7}Be is mainly the result of wet deposition by wash-out, and not by rain-out. Beryllium-7 concentration is used as an index to examine artificial radioactive contamination level.

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Sunspot number Royal Observatory of Belgum Sun spot Index Data Center(SIDC)

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