Study of the factors influence on variation of Be-7 concentration in surface air at Osaka, Japan

P Noithong¹, A Rittirong¹ and R Hazama¹

¹ Graduate School of Human Environment, Osaka Sangyo University, 3-1-1 Nakagaito, Daito, Osaka 574-8530, Japan.

E-mail: jaja.boky@gmail.com

Abstract. The concentration of ⁷Be in surface air and rainwater at Daito, Osaka (34.71 N, 135.64 E) has been measured during the period from January 8 to October 29, 2018 and April 1 to October 31, 2018, respectively. The weekly concentration of ⁷Be in surface air in the range of 8.92 ± 0.08 to 1.03 ± 0.02 mBq/m³ showing seasonal trend with maxima in spring and autumn and minima in summer, which can be ascribed to the transport of air masses from low latitudes carrying low ⁷Be concentration into the middle latitude region. The factors influences on the concentration of ⁷Be were examined. It was shown that the positive and negative correlation was found in PM2.5 (cc = 0.62) and relative humidity (cc = –0.57), respectively. The weak positive correlation was found in cosmic ray intensity (cc = 0.23), the weak negative correlation was found in precipitation (cc = –0.14) and sunspot number (cc = –0.05). The deposition rate was analyzed by using the rainwater samples. It shown negative correlation between deposition rate and precipitation (cc = –0.53). High volumes of precipitation can reduce the ⁷Be concentration of air by wash-out.

1. Introduction

Beryllium-7 (⁷Be) is a natural cosmogenic radionuclide and it is continuously produced in the upper troposphere and lower stratosphere. The production of ⁷Be in the atmosphere is mainly due to spallation reaction between galactic cosmic ray (neutron and protons) and nitrogen and oxygen. The ⁷Be then become attached to aerosols and are transported to the lower atmosphere [1]. The variation of ⁷Be concentrations in near-surface air are commonly associated with factors such as solar activity level, latitude, altitude, seasonal air mass transport, and meteorological conditions [2].

It is well known that the galactic cosmic-ray intensity at the earth’s orbit is inversely related to solar activity. So, the increase in the solar activity has accompanied a decrease in the galactic cosmic-ray intensity, which will be followed by a decrease in the production rate of cosmic-ray products, such as ⁷Be. [3]. A negative correlation is expected between the ⁷Be concentrations and sunspots. In case of particulate pollutants, since other pollutants or particulate matter (PM) are attached to aerosols, ⁷Be can be used as a tracer of atmospheric pollutants. The concentrations and variation of ⁷Be in the atmosphere [4]. The activity concentration of ⁷Be in outdoor air is about 3 mBq/m³. It will give an average yearly effective dose of 0.01 μSv worldwide [5]. The main transport route is via precipitation because of its rather short half-life (53.3 d). Since this radionuclide is easily washed out of the atmosphere by rain. The study of the relationship between precipitation and deposition very important because the rainfall rate indicating the importance of washout of the atmospheric aerosol. Thus the variation of ⁷Be concentrations varies considerably depending on the weather [4]. In addition,
variation of $^{7}\text{Be}$ concentrations relates to the areas of study, in Japan many studies report that its variation shows a seasonal pattern with a spring-autumn maximum and summer minimum [6]. Because in the summer, the wind blows and transporting air masses from low latitudes and it is well known that the air mass from low latitudes carries low concentrations of cosmogenic radionuclides [7].

This study investigates the relationship between the concentrations and variations of $^{7}\text{Be}$ in surface air and sunspot number, cosmic ray intensity, precipitation, relative humidity, and concentrations of particulate matter (PM2.5). The seasonal variation at Osaka in 2018 was studied and compared with our previous study in 2015 [8] and 2016 [9], and a long term study from 1993 to 1997 at Osaka, which was reported by Kazuko Megumi, et al. 2000 [7]. The deposition of $^{7}\text{Be}$ and its deposition rate also was studied in the area of study by using the rainwater sample.

2. Experiment
Weekly (January 8 – October 29, 2018) air samples and monthly rainfall samples (April 19 – October, 30, 2018) were collected at the rooftop of the Building No. 16 (height above ground level 31 m) located in Osaka Sangyo University, Daito, Osaka (34.71 N, 135.64 E). The air samples were collected to analyze the $^{7}\text{Be}$ concentrations. It was continuously pumped by a high volume air sampler (HV - 1000R, SIBATA) with a constant flow rate of 1000 L/min and drawn through the glass fiber (GB-100R, 203 x 254 mm², 0.3 µm pore size) and a filtered air volume of about 10080 m³. Monthly rainfall samples were collect to analyze the depositions of $^{7}\text{Be}$ and have been collected continuously with dust fall sampler (SIBATA) and collection area is 0.07 m². Pure water was poured into the dust fall sampler on the first and the last day of collecting period in the month. The collected rainfall was filtrated and evaporated to dryness in a qualitative paper filter (Advantec no. 1 ø 300 mm). The filters from air samples and rainwater were measured by HPGe detector (GX2018, CANBERRA) with 10 cm thickness of lead shield to reduce the background radiation. The detector was connected to DAQ system to record gamma ray signal of 477 keV of $^{7}\text{Be}$. Activity of $^{7}\text{Be}$ from air samples was used to calculate the surface air $^{7}\text{Be}$ concentrations in mBq/m³. Activity of $^{7}\text{Be}$ from rainwater samples was used to calculate the deposition and deposition rate in in MBq/km².month and MBq/km².mm, respectively.

The correlation between weekly variation of $^{7}\text{Be}$ concentrations and the factors influence on variation are sunspot number, which was provided by NOAA, USA [10], neutron intensity, which was provided by Cosmic Ray Station of the University of Oulu, Finland [11], PM2.5 (Particulate Matter), which were measured at Daito City Hall (34.71 N, 135.62 E) via Osaka Prefectural Government, Air Pollution constant monitoring data file [12], and precipitation and relative humidity, which were provided by our weather station at the Research Center building located in Osaka Sangyo University, Daito, Osaka (34.71 N, 135.64 E) were analyzed. Mean monthly variations of $^{7}\text{Be}$ concentration obtained from the weekly data in 2018 were compared with our previous study in 2015 [8] and 2016 [9], and the long term study at Sakai, Osaka (34.32 N, 135.30 E) from 1983 to 1997 [7].

3. Results and discussion

3.1. General features for surface air concentrations of $^{7}\text{Be}$ at Osaka
The concentration of $^{7}\text{Be}$ in mBq/m³ has been measured at our university campus in Daito, Osaka, the sampling period from January 8 to October 29, 2018. All the data are shown in figure 1. The weekly concentrations of $^{7}\text{Be}$ in surface air ranged from 8.93 ± 0.08 mBq/m³ in March to 1.03 ± 0.02 mBq/m³ in August and mean is 4.78 ± 2.00 mBq/m³.

Figure 2 shows the seasonal variation in surface air concentration of $^{7}\text{Be}$ at Daito, Osaka in 2018 and comparing the data at Daito Osaka in 2018 with our previous data in 2015 [8] and 2016 [9], and the long term data from 1993 to 1997 at Sakai, Osaka, which was reported by Kazuko Megumi, et al. 2000 [7]. Four data of the monthly average concentration of $^{7}\text{Be}$ in surface air at Osaka show the same trend and clearly show maxima in spring and autumn and minima in summer. This tendency, two-peak variation pattern, found at Osaka is almost in agreement with other observations in Japan such as Nagano [1] and Yamagata [13]. The general aspect of the seasonal variations in Japan can be interpreted that in spring and autumn, the moving high-pressure, which includes stratospheric air containing high concentrations.
of $^7$Be carried into the troposphere, passes through from West to East [1]. On the other hand, in summer the air masses are transported from the subtropical high-pressure zone or low latitudes, which are carrying low $^7$Be concentrations into the middle latitude region [7]. In each season the transient meteorological situations such as horizontal transport and moving of air masses would cause temporal changes in surface air concentrations of $^7$Be [1].

![Figure 1](image1.png)

**Figure 1.** Weekly concentrations of $^7$Be surface air during the period from January 8 to October 29, 2018 at Daito, Osaka.

![Figure 2](image2.png)

**Figure 2.** Comparing the data at Daito Osaka in 2018 (square) with our previous data in 2015 (Triangle) [8] and 2016 (circle) [9], and the long term data from 1993 to 1997 at Sakai (Pentagon) [7].

The monthly deposition of $^7$Be was observed by using rainwater samples, the sampling period from April, 19 to October, 30, 2018. The result is shown in table 1. Rainfall is believed to be the most important process affecting the deposition of $^7$Be [14]. The positive correlation was observed between the monthly deposition and the monthly precipitation with a Correlation Coefficient (cc) equal to 0.55. The increase of monthly precipitation can increase the amount of $^7$Be monthly deposition. The negative correlation was observed between $^7$Be monthly deposition rate and monthly precipitation with Correlation Coefficient equal to $-0.53$. The deposition rate was obtained by dividing the monthly deposition of $^7$Be by the monthly precipitation. The deposition rate shows decrease in accordance with
increase in monthly precipitation. Our results are in a good agreement with the long term data from 1993 to 1997 at Sakai, Osaka, which was reported by Kazuko Megumi, et al. 2000 [7]. Thus wet deposition is the predominant process in deposition and the high volumes of precipitation can reduce the $^7$Be concentration of air by washout [14].

Table 1. Amount and rate of monthly deposition and monthly precipitation.

| Month | $^7$Be monthly deposition (MBg/km²-month) | $^7$Be monthly deposition rate (MBg/km²-mm) | Monthly precipitation (mm/month) |
|-------|------------------------------------------|---------------------------------------------|----------------------------------|
| APR   | 48.30                                    | 0.88                                        | 55                              |
| MAY   | 6.21                                     | 0.03                                        | 233.4                           |
| JUN   | 17.01                                    | 0.09                                        | 183                             |
| JUL   | 78.89                                    | 0.22                                        | 366.2                           |
| AUG   | 0.69                                     | 0.01                                        | 53.6                            |
| SEP   | 56.95                                    | 0.20                                        | 290                             |
| OCT   | 29.57                                    | 0.82                                        | 36                              |
| Correlation Coefficient (cc) | 0.55 | – 0.53 | |

3.2. Correlation between weekly variations of $^7$Be concentrations in surface air and the factors influence on variation

In order to obtain information concerning the main mechanism controlling the $^7$Be surface concentration, correlation coefficients were calculated for the weekly variations of $^7$Be concentrations and relative humidity, precipitation, sunspot number, cosmic ray intensity, and PM2.5 concentration on surface air, the results are shown in figure 3.

Figure 3 (a) and (b) show the correlation between the weekly concentration of $^7$Be in surface air and relative humidity and Precipitation, respectively. A negative correlation can be observed between both parameters. The negative correlation was observed in relative humidity (cc = – 0.57) and the weak negative correlation was observed in precipitation (cc = – 0.14). Thus the relative humidity has stronger relationship with $^7$Be concentrations than precipitation. The explanation for this behavior is the high humidity leads to faster aerosol deposition, thus removing adsorbed $^7$Be from the surface atmosphere [15]. When humidity is higher, their size increases and their residence time in the atmosphere decreases under the influence of gravity, $^7$Be resulting in a lower content in the surface air [16]. The precipitation had not any influence on the levels of $^7$Be concentration in surface air and the removal of $^7$Be from the atmosphere by a strong rainfall event seems to be invalidated immediately after the event and the $^7$Be activity concentrations on surface air to be reestablished rapidly [17].

Figure 3 (c) and (d) show the correlation between weekly concentration of $^7$Be in surface air and sunspot number, cosmic ray intensity, respectively. Sunspot number was obtained from NOAA, USA [10] and cosmic ray intensity, which was obtained from the neutron-monitoring data and provided by Cosmic Ray Station of the University of Oulu, Finland [11]. It is well known that $^7$Be is a cosmogenic radionuclide and the galactic cosmic-ray intensity at the earth’s orbit is inversely related to solar activity [3]. The sunspot number is commonly used as a solar parameter related to solar activity [18]. The positive was expected in cosmic ray intensity and negative correlation was expected in sunspot number. In our results, the weak correlation was observed in both factors. The correlation coefficient equal to – 0.05 was found in sunspot number and 0.23 in cosmic ray intensity, so both factors were not significant correlation in variations of $^7$Be concentrations. However, a clear correlation was observed by long-term sampling periods. During 1983 to 1997 at Sakai, Osaka, it is 15 years sampling period was observed by Kazuko Megumi, et al. 2000 [7]. Thus, a longer sampling period should be required to confirm our results.
Figure 3. Correlation between weekly variations of $^7$Be concentrations in surface air and relative humidity (a), precipitation (b), sunspot number (c), cosmic ray intensity (d) and PM2.5 (e).

Figure 3 (c) and (d) show the correlation between the weekly concentration of $^7$Be in surface air and PM2.5 concentration. The positive correlation was observed in PM2.5 ($cc = 0.62$). We found that when the concentration of PM2.5 rises, the increasing of $^7$Be concentration was observed. The maximum value of PM2.5 concentration was found in March and the minimum value in August. This is a good agreement with the concentration of $^7$Be. The results may be caused by the concentration of PM2.5 may alter the behavior of $^7$Be. By contrast, the transport and distribution of particulate pollutants can be traced and predicted based on the behavior of $^7$Be [2]. In contrast, in the summer season, the increase of PM2.5 concentration was found but the $^7$Be concentration is low. Thus, the variation of PM2.5 concentration is not sensitive enough to vary with the seasonal variation of $^7$Be concentration. The factor of air mass direction was believed to be the most significant in the variation of $^7$Be concentration [16].
4. Conclusion

The weekly concentration of $^7$Be in surface air ranged from 8.93 ± 0.08 mBq/m$^3$ in March to 1.03 ± 0.02 mBq/m$^3$ August and mean is 4.78 ± 2.00 mBq/m$^3$ showing seasonal trend with maxima in spring and autumn and minima in summer, which can be ascribed to the transport of air masses from low latitudes carrying low $^7$Be concentration into the middle latitude region. The positive correlation was observed in the monthly deposition (cc = 0.55) and the negative correlation in the $^7$Be monthly deposition rate (cc = −0.53) with the monthly precipitation. We found that wet deposition is the predominant process in deposition and the high volumes of precipitation can reduce the $^7$Be concentration of air by washout. We found that the relative humidity (cc = −0.57) has stronger relationship with $^7$Be concentrations than precipitation (cc = −0.14). The higher relative humidity leads to increasing the aerosols size and their residence time in the atmosphere decreases under the influence of gravity. Sunspot number and neutron intensity were not significant correlation in variations of $^7$Be concentrations in our study. The positive correlation was observed in PM2.5 (cc = 0.62). The concentration of PM2.5 can alter the behavior of $^7$Be but it is not sensitive enough to vary with the seasonal variation of $^7$Be concentration.

5. References

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