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Deposition of beryllium-7 in Hsinchu, Taiwan

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ABSTRACT

In the present study, factors that influence the distribution and variation of ⁷Be in Hsinchu, Taiwan were elucidated. The ⁷Be activity including the deposition flux and air concentration was continuously monitored and recorded throughout a 15-year period (1996–2010). To explain the observed variability in the ⁷Be activity over time, air concentration and deposition flux of ⁷Be were correlated to rainfall and solar activity. The monthly average deposition flux and air concentration of ⁷Be were inversely related to solar activity with the 11-year cycle and were not strongly correlated to rainfall. The highest seasonal deposition flux of ⁷Be occurred in March, which is commonly referred to as the spring maximum, due to air-mass mixing processes in the troposphere. The air concentration of ⁷Be was seasonally variable and was significantly affected by monsoons. The lowest deposition flux and air concentration of ⁷Be were observed in July and August due to the occurrence of southwest monsoons from low latitudes, which carry air masses with low concentrations of ⁷Be. The deposition flux was enhanced by precipitation, which increased the deposition velocity, transferring more ⁷Be from the troposphere to the ground. The fraction of dry to total deposition varied seasonally and was equal to 9.86%, on average.

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1. Introduction

Beryllium-7 (⁷Be) is a naturally occurring radionuclide produced by spallation reactions through interactions of galactic cosmic rays with nitrogen and oxygen in the stratosphere and upper troposphere. Production rates of ⁷Be and other cosmogenic radionuclides have been simulated in the earth's atmosphere to determine their variation with latitude, solar activity and geomagnetic field intensity (Masarik and Beer, 1999; Nagai et al., 2000; Usoskin and Kovaltsov, 2008). The sunspot number is an indicator of solar activity, which can enhance the magnetic field surrounding the earth and reduce the intensity of cosmic rays and production rates of cosmogenic radionuclides in the atmosphere. After ⁷Be is formed, it rapidly associates with submicron-sized aerosol particles (Bondietti et al., 1984). The concentration of ⁷Be in the troposphere is altitude and latitude dependent and is equal to 12.5 mBq m⁻³, on average (UNSCEAR, 2000).

The concentration of ⁷Be in the near-surface environment has been measured to explain its variation with respect to solar activity, rainfall, latitude and meteorological factors. The intensity of galactic cosmic rays is modulated by solar activity, thus, the ⁷Be concentration of the air changes over an 11-year cycle (Azahra et al., 2003; Kulan et al., 2006; Papastefanou and Loannidou,

2004; Kikuchi et al., 2009). The concentrations of ⁷Be in the surface air and rainwater have been frequently used to estimate the deposition flux of ⁷Be, which is affected by both dry and wet processes. In wet deposition, precipitation transports ⁷Be from the upper troposphere to the ground. This process usually occurs in the spring and summer, when air transport from the stratosphere to the troposphere is easily induced by the heating of the earth's surface. Positive correlations between precipitation (rain or snow) and the deposition flux of ⁷Be have been observed in previous studies (Ayub et al., 2009; Doering and Akber, 2008; Gonzalez-Gomez et al., 2006; Harvey and Matthews, 1989; Ishikawa et al., 1995).

The deposition velocity of aerosols can be affected by precipitation and seasonal factors. However, the deposition velocity reflects the effectiveness of aerosol scavenging by precipitation; thus, both the amount of precipitation and duration of rainfall must be considered. Rainfall reduces the air concentration of ⁷Be (Arkian et al., 2010; Kikuchi et al., 2009) and increases the deposition velocity, which provides important information on the behavior of atmospheric particulates in the environment (Garger, 1994; Young and Silker, 1980). The fraction of dry to total deposition of ⁷Be, which is dependent on regional climatic conditions, is experimentally estimated to be approximately 10% (Ioannidou and Papastefanou, 2006; Rosner et al., 1996; Simon et al., 2009; Todd et al., 1989).

The production rate of ⁷Be in the atmosphere increases with a decrease in latitude, while the maximum deposition flux occurs in

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the middle latitudes due to vertical/horizontal air mass transport and the frequency of rainfall events (Kulan et al., 2006). In the summer, air masses from low latitudes carrying low concentrations of ^7Be dilute the surface air in Japan (Feely et al., 1989; Megumi et al., 2000). In Taiwan, the southwest monsoon is predominant in the summer time, and the northeast monsoon is predominant in the winter (Chen and Chen, 2003; Kerns et al., 2010). The northeast and southwest monsoon are accompanied by significant amounts of rainfall, which may alter the distribution of ^7Be .

In the present study, the deposition flux and air concentration of ^7Be were simultaneously monitored (monthly data) from 1996 to 2010. All of the monthly data were correlated to the sunspot number and precipitation to identify factors affecting the deposition and concentration of ^7Be in Hsinchu, Taiwan. The effects of monsoons, precipitation and the rainy season on the deposition flux and air concentration of ^7Be were considered. The deposition velocity of ^7Be was determined and was associated with precipitation to estimate the deposition flux from dry and wet processes.

2. Materials and methods

The activity of ^7Be in Hsinchu, Taiwan, including the deposition flux and air concentration was monitored from 1996 to 2010. During this period, the average annual precipitation of the study region was equal to 1800 mm. Deposition and air samples were collected from the building roof of the Radioactivity Measurement Laboratory of National Tsing Hua University (24 47' N, 120 59' E, altitude=30 m).

2.1. Sample preparation

Wet and dry deposition samples were obtained using a large-area deposition collector (1 m $L \times$ 1 m $W \times$ 0.30 m H). Fallout samples were collected on a monthly basis and were dried to reduce the sample volume. Subsequently, radiometric analysis was conducted to determine the deposition flux of ^7Be ($\text{Bq m}^{-2} \text{s}^{-1}$). Air samples were collected once a week with a glass fiber filter (47 mm in diameter) mounted on a high-volume air sampler (HD-28, RADeCO) set at a flow rate of 40 LPM. Filters collected on a monthly basis were combined and were subjected to gamma-ray counting to determine the ^7Be concentration of the surface air (Bq m^{-3}).

2.2. Gamma-ray counting

Deposition and air samples were counted with a germanium detector (GC3520; Canberra) connected to a multi-channel analyzer (MCA 35-plus; Canberra). The activity of ^7Be was determined by following the emission of gamma rays at 478 keV and the spectra were analyzed with SAMPO 90 software. The ^7Be activities were decay-corrected to the mid-point of sample collection.

2.3. Related records

The volume (mm) and duration (%) of rainfall and the number of raining hours per month in Hsinchu were collected from the Central Weather Bureau of the Republic of China (Taiwan). The monthly duration of rainfall was calculated based on hourly records. The sunspot numbers were provided by Solar Influences Data Analysis Center (SIDC, <http://sidc.oma.be/sunspot.data/dailyssn.php>).

2.4. Deposition flux and velocity

In previous studies, both the deposition flux and air concentration were utilized to determine the deposition velocity (V) of ^7Be (Garger, 1994; Kulan et al., 2006; Young and Silker, 1980), as expressed by the following equation:

$$F = CV \quad (1)$$

where F is the deposition flux of ^7Be ($\text{Bq m}^{-2} \text{s}^{-1}$) and C is ^7Be concentration of the air (Bq m^{-3}). Both the deposition flux and air concentration of ^7Be were measured in the present study.

3. Results and discussion

3.1. Variation of ^7Be with solar activity

In Fig. 1 the deposition flux and air concentration of ^7Be are plotted together with the sunspot number observed during 1996–2010. The lowest deposition flux and air concentration of ^7Be occurred during 2000–2002, when the maximum solar activity of the 23rd solar cycle was observed. Similar results obtained during this time frame have been reported in other areas (Kikuchi et al., 2009; Kulan et al., 2006). The production rate of ^7Be in the atmosphere is influenced by the solar activity, or the intensity of cosmic rays. However, the strength of this effect decreases with an increase in latitude and altitude (Masarik and Beer, 1999; Kikuchi et al., 2009).

3.2. Effect of precipitation on the deposition and air concentration of ^7Be

Rainfall is believed to be the most important process affecting the deposition of ^7Be . However, the contribution of precipitation to ^7Be deposition is difficult to discriminate from other factors. The deposition flux of ^7Be did not significantly vary with rainfall, and considerable changes in the deposition flux were not observed during heavy rainfall events (Fig. 2(a)). More than 80% of the monthly deposition fluxes were less than $5 \times 10^{-5} \text{Bq m}^{-2} \text{s}^{-1}$, and a weak correlation ($r^2=0.043$) with precipitation was observed (Fig. 2(b)). Compared to the volume of precipitation, a stronger correlation ($r^2=0.20$) with the duration of rainfall (%) was detected (Fig. 2(c)). Thus, high deposition fluxes may be attributed to seasonal factors, as described in the following section.

Typically, the ^7Be concentration of air is inversely related to rainfall. High volumes of precipitation can reduce the ^7Be concentration of air by washout (Fig. 3(a)). However, in the present study, precipitation and rainfall duration showed insignificant correlations ($r^2 < 0.02$) with the ^7Be concentration of air (Fig. 3(b) and (c)). The average monthly air concentrations of ^7Be was equal to be 5.66mBq m^{-3} , and should be characterized according to latitude in Taiwan.

3.3. Seasonal factors affecting the ^7Be concentration

The monthly average deposition flux and air concentrations of ^7Be throughout the entire annual cycle was related to the volume of rainfall. However, as illustrated in Fig. 4(a) and (b), seasonal variations in the ^7Be concentration did not correspond to the rainfall pattern. Similar to the results of previous studies, the highest deposition of ^7Be was observed in March, which is commonly referred to as the spring peak (Arkian et al., 2010; Dibb, 1989; Megumi et al., 2000; Yoshimori, 2005). During this time frame, stratospheric air intrudes into the troposphere, introducing ^7Be to the troposphere (Feely et al., 1989; Simon et al., 2009). In northern Taiwan, tropopause reaches its annual maximum in winter and spring (Feb–May), and a minimum in summer and autumn (June–Nov) (Thulasiraman et al., 1999; Das et al., 2009).

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