

Be-7 MEASURED AT GROUND AIR LEVEL AND RAINFALL IN THE CITY OF SÃO PAULO

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ABSTRACT

The cosmogenic radionuclide ${}^7\text{Be}$ ($T_{1/2} = 53.3$ d), produced in the upper atmosphere by cosmic ray spallation of oxygen and nitrogen, is one of the cosmogenic radionuclides that can be used as tracer for heavy metals and pollutants in the environment, tracer of soil erosion and sedimentation in lakes, among other examples. Their subsequent deposition to the land surface occurs as both wet and dry fallout, although it has been demonstrated that ${}^7\text{Be}$ fallout is primarily associated with precipitation. There is limited data on the concentration of ${}^7\text{Be}$ in rainfall and in particulate in the Southern Hemisphere and in Brazil, compared with data from the Northern Hemisphere. This paper presents the results obtained of ${}^7\text{Be}$ concentrations measured from April 2011 to June 2013, in samples of air at ground level, each fifteen days, and rainfall in all the rainy events that occurred at Instituto de Pesquisas Energéticas e Nucleares (IPEN), which has its campus located in the city of São Paulo, state of São Paulo, Brazil. The concentrations of ${}^7\text{Be}$ were measured by non-destructive gamma-ray spectrometry using an extended range closed-end coaxial Be-layer HPGe detector with 25% relative efficiency and associated electronic devices and live counting time varying from 100,000 s to 300,000 s. The results obtained of ${}^7\text{Be}$ in particulate and in rainfall were correlated to seasons, precipitation, temperature and sunspot number. The higher values obtained for the concentrations were in spring and summer time presenting good correlations with the amount of precipitation and sunspot number and a clear seasonal variation.

1. INTRODUCTION

${}^7\text{Be}$ ($T_{1/2} = 53.3$ days), a natural radionuclide of cosmogenic origin, is continuously produced in the upper atmosphere by the spallation process of light atmospheric nuclei such as nitrogen and oxygen when they interact with protons and neutrons of cosmic rays. These atoms readily react to produce BeO or $\text{Be}(\text{OH})_2$ molecules, which in turn diffuse through the atmosphere until they attach to an atmospheric aerosol. Subsequent deposition to the land surface therefore occurs as both wet and dry fallout, although it has been demonstrated that ${}^7\text{Be}$ fallout is primarily associated with precipitation [1].

This cosmogenic radionuclide is used as tracer for heavy metals and pollutants in the environment [2], for tracing soil erosion [3], transport processes in watershed and chronometer in aquatic and atmospheric systems lakes [1, 4-5]. The study of these processes requires a good knowledge of its input from the atmosphere, and the ${}^7\text{Be}$ behavior study in the atmosphere will give insight on the behavior of similar chemical species in atmosphere aerosols.

About 70% of the ^7Be is produced in the stratosphere and the remaining 30% in the troposphere. In general, most of the ^7Be do not reach the troposphere except for spring when season thinning of the troposphere occurs at mid-latitudes, resulting in air exchange between stratosphere and troposphere [5]. The ^7Be produced in the stratosphere usually stays there for about a year before entering the troposphere, where it remains for about 6 weeks. Its transfer to Earth's surface is achieved by gravitational settling and precipitation. The study of possible seasonal changes in ^7Be concentrations due to an increase of its migration from the stratosphere into the troposphere gives useful information about its distribution and behavior [6].

Cosmic radionuclide concentration in air at ground level can vary greatly according to latitude and altitude, season of the year and local meteorological conditions. ^7Be concentration in surface air depends on wet scavenging, stratosphere to troposphere exchange, downward transfer in the troposphere and horizontal transfer from middle and subtropical latitudes to higher and lower latitudes [7, 8]. Generally the values measured at middle latitudes are higher than those at northern latitude [6, 9-10]. The production rate of ^7Be changes according to variations in flux of cosmic rays caused by the 11-year sunspot cycle. This phenomenon is one of the major parameters that cause the variation in ^7Be concentration in surface air [11, 12].

The aim of this paper is to present preliminary results of the ^7Be concentration in ground level air and rainfall in the city of São Paulo, SP, Brazil. From April 2011 to June 2013, samples of particulate and rainfall have been measured by gamma spectrometry and the resulting seasonal ^7Be data were correlated to seasons, rainfall, temperature and sunspot number.

2. MATERIALS AND METHODS

2.1. Site Description

The sampling site is located at IPEN – Instituto de Pesquisas Energéticas e Nucleares, which is approximately 10 km west from downtown São Paulo. The city of São Paulo is situated on a plateau in southeastern Brazil, at latitude 23°33'58.27"S and longitude 46 44'14.82"W, with an average altitude of 760 m above sea level. Climate in the area is temperate tropical with dry period in winter and rainy in summer. The annual average temperature for the studied period was 21.4°C, showing minimum and maximum average temperature of 16.9°C and 25.8°C, respectively. The annual rainfall in IPEN campus, for the studied period, ranged from 1590 mm to 2081 mm and the winds are predominantly from the SSE sector.

2.2. Sampling

In the framework of an environmental radiological monitoring program carried out at IPEN facilities, due to their routine gaseous effluents releases, atmospheric air is sampled at 1.0 m above the ground. The air is continuously pumped by a low volume sampler (Staplex model LVC-50) and forced to pass through cellulose filters with 47 mm diameter with retention efficiency near 100%, for the collection of particulates. The filters are replaced every 15 days, which correspond to a filtered air volume of about 3,000 m³.

Rainfall was collected in each rainy event from April 2011 to June 2013, in a polyethylene container of 0,132 m², totaling 123 rainy events. At the end of each rainy event and when the sample was collected, the container was rinsed with 300 mL of 1M HNO₃ and 200 mL super pure water, which were added to the sample. It was only considered precipitation values greater than 5 mm, because this volume is used to characterize different radionuclides deposition process [4, 13]. The amount of precipitation in mm was obtained from a rain gauge installed at the sampling point. Since the rainfall container was exposed to the atmosphere continuously, it was collected the total deposition, that is, the wet and dry fallout. All the collected samples were acidified with HNO₃ at pH lower than 2, in order to avoid losses due to adsorption on the concentration process, and then concentrated to a final volume of 100 mL.

2.3. Be-7 determination

Measurement of ⁷Be, in both samples, particulate and rainfall, was carried out by non-destructive γ -ray spectrometry using the 477.61 keV γ -ray. An extended range closed-end coaxial Be-layer HPGe detector with 25% relative efficiency, 2.09 keV resolution at 1.33 MeV and associated electronic devices was used, with live counting time ranged from 100,000 to 300,000 s. The spectra were acquired by multichannel analyser Ethernim and, for the analysis, WinnerGamma software was used. The activity was corrected for decay of ⁷Be to the date of collection. The associated uncertainty for one sigma confidence ranged from 10% to 30% from the obtained results.

3. RESULTS AND DISCUSSION

3.1. ⁷Be in particulate

Fig. 1 presents the ⁷Be results, in mBq m⁻³, plotted in function of the sampling date for the study period (April 2011 to June 2013); the values ranged from 0.31 ± 0.07 mBq m⁻³ in September 2012 to 2.44 ± 0.15 mBq m⁻³ in March 2013, winter and summer time, respectively.

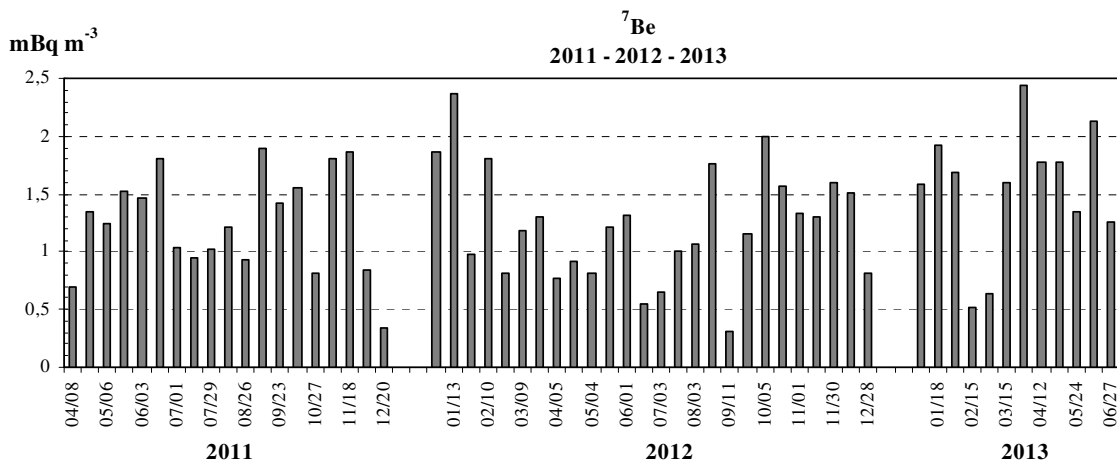


Figure 1. ⁷Be in mBq m⁻³ in particulate for the study period (April 2011 to June 2013).

Fig. 2 presents the correlation of the average monthly ^7Be concentration, in mBq m^{-3} , in particulate with sunspot number, for the study period. A good correlation, inverse relationship, but not totally perfect was observed. This inverse correlation is attributed to the cyclic pattern resulting from about 11-year temporal variations associated with the solar activity, which has a direct relation with a sunspot number. The present work started in the maximum of the cycle 24, which began in 2008 and had a peak in November 2011, with the highest sunspot number. From this month forth, the sunspot number presented a trend of decrease, what can be observed in Fig. 2. The sunspot numbers were obtained from the Sunspot Index Data Center (SIDC) in Brussels [14].

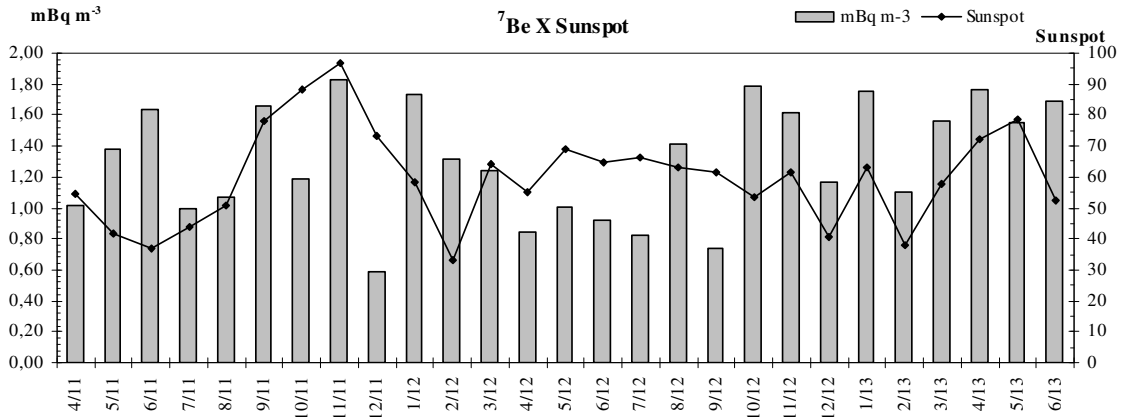


Figure 2. Correlation between ^7Be , mBq m^{-3} in particulate and sunspot number.

Fig. 3 presents the correlation of the average monthly ^7Be concentration, in mBq m^{-3} , in particulate with average temperature in the city of São Paulo, in $^{\circ}\text{C}$, for the study period. It can be observed that, in general, there is a good correlation between ^7Be and temperature, with increase of the concentration in warmer months and decrease in colder months. The temperature data were obtained from CIIAGRO [15].

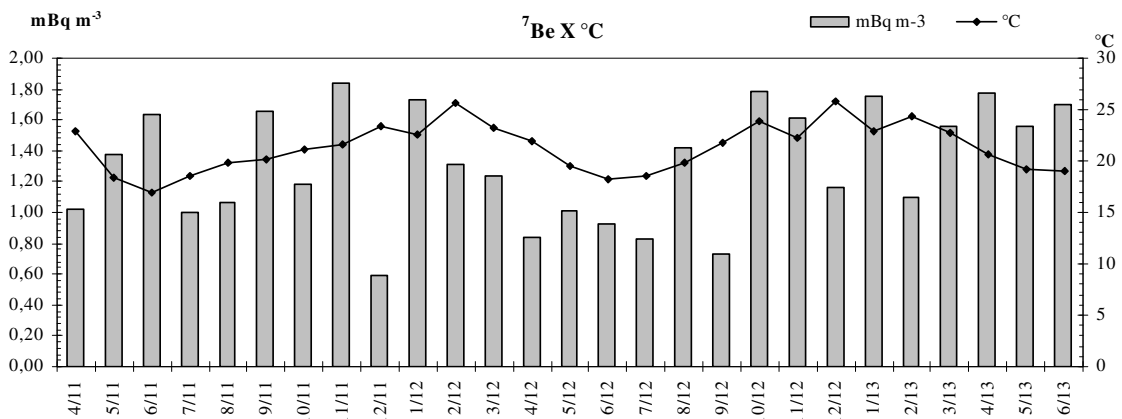


Figure 3. Correlation between ^7Be , mBq m^{-3} , in particulate and temperature

Fig. 4 presents the correlation of the average monthly ^7Be concentration, in mBq m^{-3} , in particulate with average precipitation, in mm, measured inside of IPEN campus for the study period. For the whole period studied it was found that the highest concentrations of ^7Be were obtained in months with high indexes of rainfall due to the washout of the atmosphere by the precipitation.

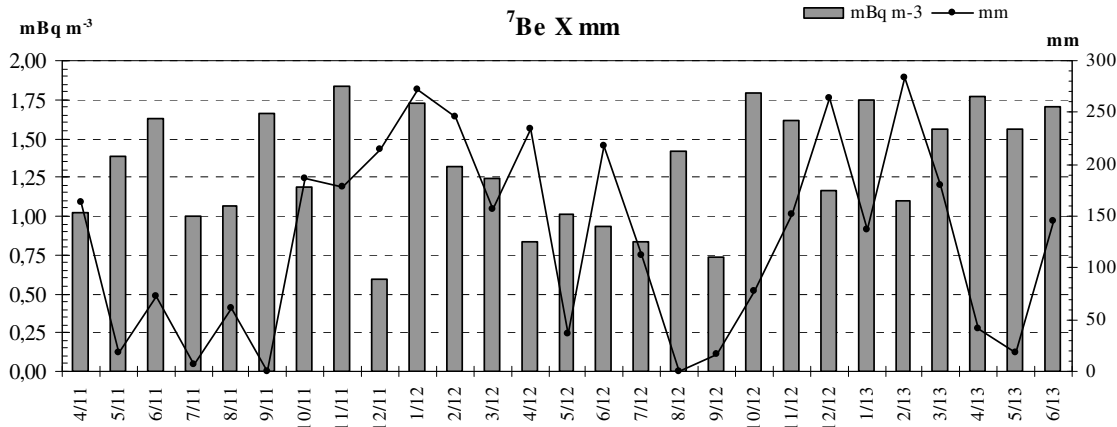


Figure 4. Correlation between ^7Be , mBq m^{-3} , in particulate and precipitation

3.2. ^7Be in rainfall

The monthly amount of precipitation for the studied period is plotted in Fig. 5. The precipitation for the study period ranged from 5 to 160 mm of each rainy event and in September of 2011 and August of 2012 there was no precipitation.

Comparing the amount of precipitation among the 26 months of study, it is possible to verify the climate of temperate tropical that is characteristic of São Paulo with dry periods in winter and rainy in summer only in 2011, where the winter months presented the lowest values of rainfall. In 2012, June and July presented high values, comparable to the summer months of January and February. In 2013, July also presented high values, like in 2012.

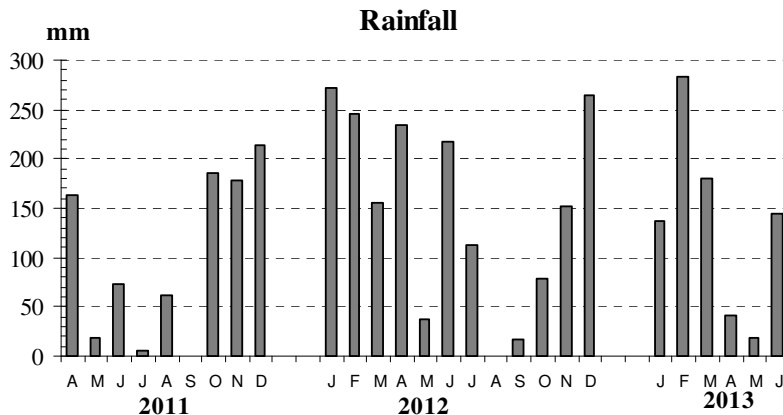


Figure 5: Monthly precipitation during the sampling period (April 2011 to June 2013).

Fig. 6, 7 and 8 present the correlation between ^7Be concentration, in Bq L^{-1} , and precipitation for 2011, 2012 and 2013, respectively. For the entire studied period it is possible to verify that there is an evident correlation of ^7Be concentration in rainfall and the amount of precipitation. The highest values obtained were found in months where the precipitation was low and inversely the lowest in months of high indexes of rainfall, due to the dilution effect of the sample [16].

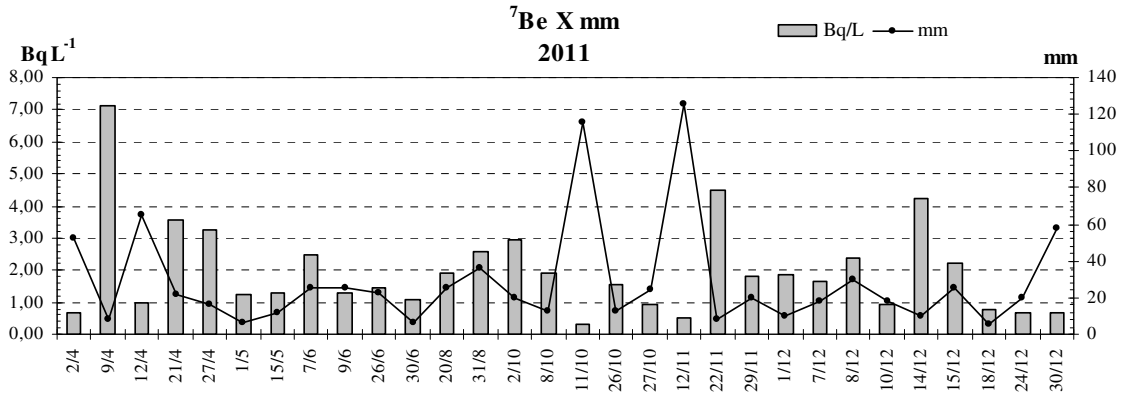


Figure 6. Correlation between ^7Be concentration, in Bq L^{-1} , and precipitation for 2011

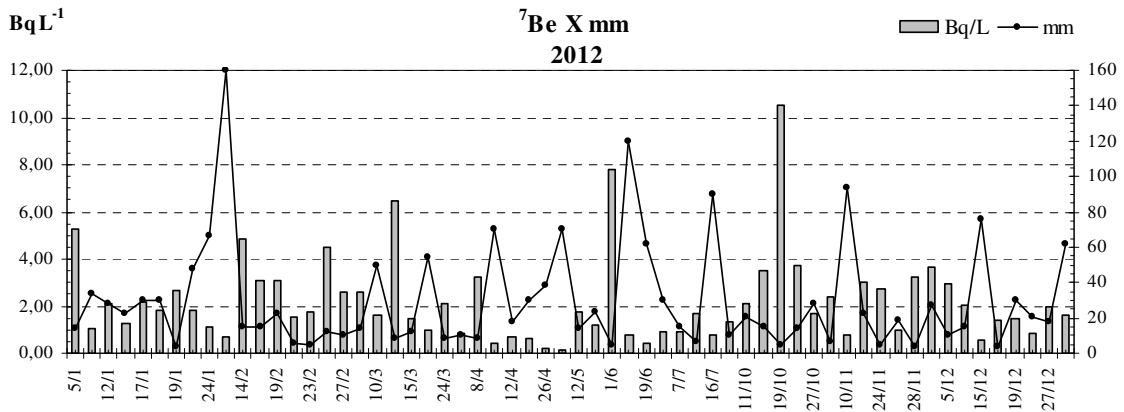


Figure 7. Correlation between ^7Be concentration, in Bq L^{-1} , and precipitation for 2012

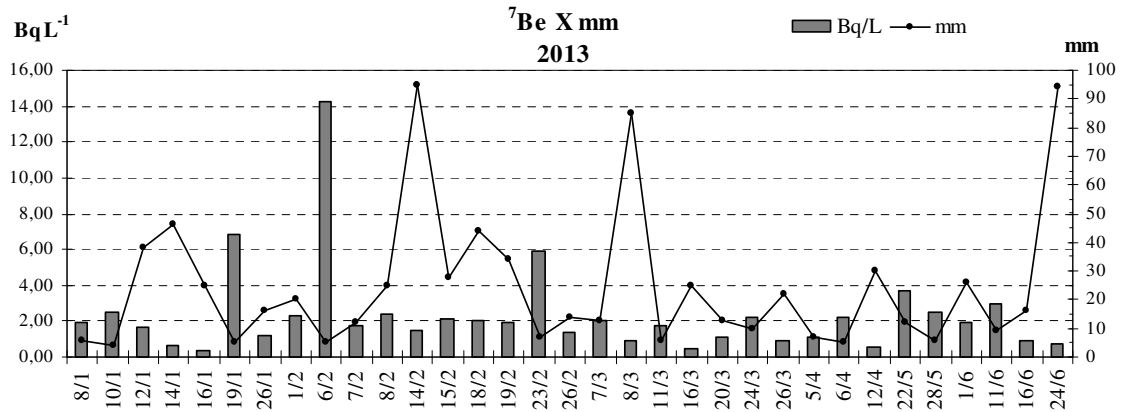


Figure 8. Correlation between ^7Be concentration, in Bq L^{-1} , and precipitation for 2013

4. CONCLUSIONS

This paper presented the preliminary results for ^7Be concentration measured at ground air level and in all the rainy events that occurred from April 2011 to June 2013, at IPEN, São Paulo, SP, Brazil in order to provide results of its concentration in Brazil and to understand the atmospheric behavior of this radionuclide in the Southern Hemisphere.

The obtained results for ^7Be concentration in air surface showed a good agreement with data from Northern hemisphere of similar latitude, in spite of the city of São Paulo being situated in low latitudes of the Southern hemisphere. ^7Be concentrations displayed clearly seasonal variations, due to the increase of its concentration during spring time and variation in production rate of ^7Be ; which is attributed to variations in the flux of cosmic rays.

For rainfall the results showed that there is an evident correlation of ^7Be concentration with the amount of precipitation, therefore high values were obtained in dry months and low values in rainy months, due to the dilution effect of the sample.

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