J Radioanal Nucl Chem (2015) 303:2151–2158
DOI 10.1007/s10967-014-3737-2

Study of the cosmogenic factors influence on temporal variation of ^{7}Be air concentration during the 23rd solar cycle in Málaga (South Spain)

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Received: 30 July 2014 / Published online: 2 November 2014

Abstract Atmospheric ^{7}Be activity concentrations were measured in Málaga (South Spain) during the period 1997–2007. Monthly concentrations of ^{7}Be ranged between 2.47 and 8.12 mBq m^{-3} showing seasonal trend with maxima in spring–summer time. Influence of solar activity, cosmic rays and aerosol optical depth on airborne ^{7}Be was analyzed. Weak negative correlation between sunspot number and ^{7}Be is found. Instead, both solar energetic protons >100 MeV and cosmic rays show positive weak significant correlation with this radionuclide. Meteorology may contribute to the low correlation coefficients found. Aerosol optical depth exhibits significant correlation with ^{7}Be, confirming that this radionuclide attaches to atmospheric particles.

Keywords ^{7}Be · Solar cycle · Sunspot number · Solar energetic protons · Cosmic rays · Aerosol optical depth

Introduction

Atmospheric ^{7}Be has been widely studied at different places around the world, like Europe [eg. 1, 2], America [3], or Asia [4, 5]. This radionuclide is a potentially useful tracer of aerosols due to its short half-life ($T_{1/2} = 53$ days), ease of measurement and well-defined source term. Most of ^{7}Be is produced in the lower stratosphere and slowly transported to the surface. ^{7}Be plays a role of atmospheric tracer and its measurements provide an important clue on atmospheric air mass motions. Thus, several works have used the radionuclide to analyze vertical transport of air masses of stratospheric origin, atmospheric long-range transport, as well as determine the air-sea exchange process [6–8]. Others have focused on estimating soil erosion rates using ^{7}Be [9, 10].

^{7}Be is a natural cosmogenic radionuclide produced in the upper atmosphere when cosmic-ray-produced neutrons and protons interact with the nuclei of nitrogen and oxygen and disintegrate them into lighter fragments in a process known as spallation [11–14]. Recent research points out the photo-nuclear reaction as another possible mechanism of ^{7}Be formation in the upper atmosphere [15]. Once the radionuclide is formed, it is attached quickly to aerosols with aerodynamic diameter between 0.4 and 2 μm [16–20].

Previous studies [3, 21–24] have reported ^{7}Be production rate has altitude dependence because of the atmospheric attenuation of the radiation (around 70 and 30 % of ^{7}Be is produced in the stratosphere and in the troposphere respectively), and latitude dependence due to the deflect effect of the magnetic field of the Earth (increasing at high latitude). Moreover, variations of the ^{7}Be concentration in the air show a seasonal pattern with a spring–summer maximum and an autumn–winter minimum [e.g. 2, 3].

Thus, galactic cosmic rays and solar energetic particles entering the heliosphere are affected by interplanetary magnetic field and solar activity [13, 14, 22, 25]. In addition, meteorological parameters such as precipitation

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Fig. 1 Location of the study area

scavenging, air exchange between the stratosphere and the troposphere and vertical transport influence on atmospheric ^{7}Be concentrations [1, 3, 5, 24, 26–29].

The aim of the present study was to investigate the effect of the solar activity, cosmic rays and the presence of airborne particulate matter on the variation of airborne ^{7}Be radioactivity surface levels during a complete solar cycle.

Materials and methods

Study area

The sampling site is located in the north-west of the city of Málaga, approximately 5 km from the coastline, close to the airport and surrounded by roads with traffic exhaust (Fig. 1). Málaga is the major coastal city of the Andalusia region, South Spain, with a population of approximately 570,000 inhabitants. The city lies on the Mediterranean coast and it is bordered to the north by a high mountain range. Thus, Málaga is affected by a Mediterranean climate with dry and hot summers, temperate winters, scarce rainfall with mean annual precipitation of around 550 mm year^{-1}, and a well developed land-sea breeze regime [30].

Sampling procedure

In the period January 1997–December 2007, 132 aerosol samples were collected weekly on the roof of a three-story building on the Faculty of Sciences of the University of (4.49°W; 36.73°N) at a height of 10 m above the ground to minimize the resuspension from soil. Aerosol samples were collected in cellulose membrane filters with a pore size of 0.8 μm and a diameter of 47 mm diameter as part of a national monitoring programme coordinated by the Consejo de Seguridad Nuclear. The collection efficiency amounted to 99.99 % with an air sampler (Radeco, mod AVS-28A) at a flow rate of 40 l/min. A monthly composite sample containing 4–5 filters was formed (average volume of 1,600 m^{3}) for the ^{7}Be determination. Measurements by high-resolution gamma-ray spectrometry were performed to determine the ^{7}Be activities of the samples using a coaxial-type germanium detector (Canberra Industries Inc., USA). The peak analysis of ^{7}Be (I = 10.52 %, 477.7 keV) was done using SPECTRAN at peak analysis software. The spectra acquisition time was 172,800 s. Details of the low-background gamma-ray detection system used and the calibration of the resulting samples for gamma spectrometry have been previously described by Dueñas et al. [27, 31].

Table 1 Descriptive statistical parameters of monthly ^{7}Be activity concentrations (mBq m^{-3})

	AM	GM	SD	SK	Max	Min	CV (%)
^{7}Be	4.74	4.61	1.12	−0.04	8.12	2.47	23.67

Data sets

In order to describe the 11-year cycle of the solar activity, the monthly sunspot number and the Solar energetic proton (SEP) obtained from NOAA's Space Weather Prediction Center (SWPC) were used. Shielding effect was shown using magnetic field data measured by various spacecraft near the Earth's orbit. These data are provided by the Space Physics Data Facility (SPDF) at the Omni2 directory. Cosmic ray data were obtained from the neutron monitor belonging to Sodankyla Geophysical Observatory of the University of Oulu in Finland. Finally, atmospheric particulate matter was indirectly derived from the Aerosol Optical Depth measured by the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) Deep Blue Level 3 operated by NASA.

Results and discussion

^{7}Be concentration levels in surface air

^{7}Be in ground level air over a 11-year period, was measured in Málaga (Spain). Table 1 gives the summary of the descriptive statistics such as arithmetic mean (AM), geometric mean (GM), standard deviation (SD), skewness (SK), maximum and minimum values and the coefficient of variation (CV). The activity concentrations of the ^{7}Be ranged between 2.47 and 8.12 mBq m^{-3}, with an average of 4.71 ± 1.13 mBq m^{-3}. These values are coincident with those measured in the regions of similar latitude reported by Chae et al. [5] at Daejeon (36°N), Piñero-García et al. [32] at Granada (37°N), or by Narazaki and Fujitaka [33] at Dazifu (33°N).

The frequency distribution of the concentrations of ^{7}Be is illustrated in Fig. 2. As seen in this figure, the plot of ^{7}Be specific activity measured in aerosols at surface during this period showed skewed histogram which fits well with a log-normal distribution. The goodness of the fit was tested using the Kolmogorov–Smirnov test with a confidence level of 90 %.

Annual and monthly variation of ^{7}Be

Inter and intra-annual variability was expected and observed. Atmospheric ^{7}Be was below average in 2000 and 2004, close to average in 1999, 2001, 2002 and 2003, and above average in

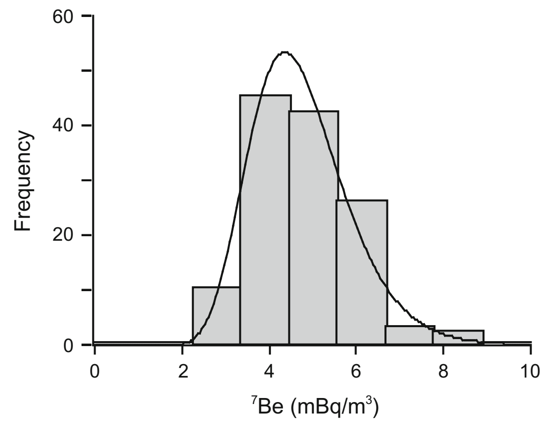

Fig. 2 Frequency histogram for monthly ^{7}Be atmospheric concentrations for the data collected from 1997–2007. *Black line* represents lognormal fitting

1997, 1998, 2005, 2006 and 2007 (Fig. 3a). These above average values correspond with the initial and final phases of the 23rd solar cycle. The significance of the differences in ^{7}Be concentrations among the different years was assessed by the corrected Mann–Whitney test. The lowest concentrations of ^{7}Be were found during the year 2000 (corresponding to the solar maximum activity), showing significant differences with the start or the end period of the solar cycle (1997, 1998, 2006 and 2007), when the highest concentrations are registered. Piñero-García and Ferro-García [34] for example, found that during the ascending phase of the solar cycle, solar activity played an important role in the production rate of ^{7}Be, whereas during the descending phase, the role of the solar activity was secondary. Moreover, concentrations of ^{7}Be show seasonal pattern with maximum values being observed in the spring–summer months and minimum in the autumn–winter months (Fig. 3b), though this variation is weaker than the one found in other studies (e.g. [2, 35]). This behaviour is representative of middle latitudes, such Málaga, due to their cosmogenic origin and the characteristics of the sampling point. Several authors have attributed the observed seasonality to a combination of the precipitation scavenging which removes from the atmosphere the aerosols where ^{7}Be is attached, or the stratosphere-to-troposphere exchange and vertical mixing within the troposphere [1–3, 29, 36, 37]. During warm months, the troposphere is less stable due to the solar heating of the earth's surface, which leads to upward transport of tropospheric species and hence a downward transport of stratospheric air parcels because of the convective movements.

Influence of solar activity in ^{7}Be variability

The solar activity modulates the heliospheric condition, which modulates the cosmic ray flux, which in turn modulates the cosmogenic isotopic production. Therefore, as ^{7}Be is a cosmogenic isotope primarily produced by nuclear

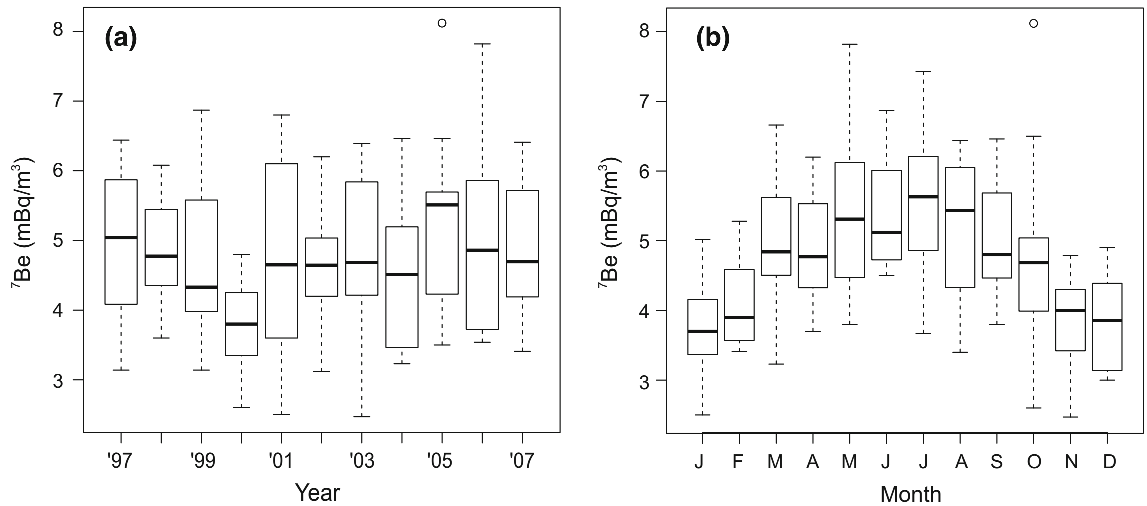

Fig. 3 **a** Boxplot of the variation in ^{7}Be activity on a yearly basis. **b** Boxplot of the mean monthly atmospheric concentrations of ^{7}Be averaged over the study period

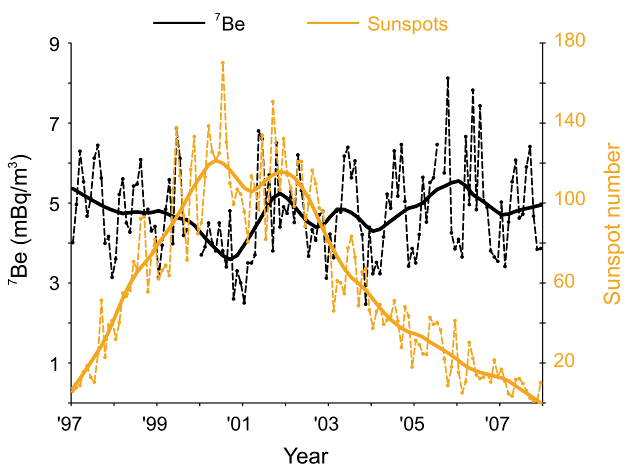

Fig. 4 Monthly concentrations of atmospheric ^{7}Be and sunspot number. *Dash-dotted lines* indicate experimental values and *solid lines* show trend component

reactions of cosmic ray and solar energetic particles with atmospheric gases such nitrogen and oxygen, ^{7}Be concentrations in surface air are affected by solar activity [38].

The sunspot number is commonly used as a solar parameter related with the solar activity [39]. Figure 4 shows the monthly average values of ^{7}Be specific activity and sunspot number. Solar activity reaches the maximum during the 2000, while ^{7}Be atmospheric concentration has the lowest values. Many studies have found that ^{7}Be concentrations are strongly inverse correlated with sunspot activity (e.g. [1, 2, 4, 35]). In the present study monthly ^{7}Be activity is not well correlated with the sunspot number and a very weak Pearson correlation coefficients ($r = -0.14$, p value $= 0.110$) was found. The weak inverse correlation observed was also found by Chae et al. [5] and Piñero-García et al. [32], who stated that meteorological factors could reduce this correlation.

The time series have four components: trend, cyclical, seasonal and irregular [40, and references therein]. The first one corresponds to the long term pattern, and the last one represents the unpredictable component, i.e. the meteorological factors. We have decomposed our data in these components. When the correlation between the trends of ^{7}Be and sunspot number is made, a statistically significant anti-correlation is found ($r = -0.59$, and p value $= 0$). Therefore, that means that when removing the seasonal and irregular components, the relation between both variables increases and hence, the weak inverse correlation of the observed data may be elucidated by meteorological factors.

When performing that correlation on a seasonal basis, it is found that summertime has the highest correlation coefficient ($r = -0.63$), while winter, spring and autumn periods do not show a significant correlation. It is noteworthy that the correlation between trend components of ^{7}Be and sunspots during the summer does not show an appreciable increase, whereas for the rest of seasons this correlation increases reaching a significant correlation. That could mean that the most apparent factor contributing to ^{7}Be variations in summertime is the solar activity while meteorological factors have greater influence on the radionuclide variations during the rest of the year.

Solar activity is associated also with the onset of solar energetic particles, both by coronal mass ejection (the weakest events), and by prompt events originating on the disk linked to flares. The geosynchronous operational environmental satellite (GOES) provides energetic particle population at geostationary orbit. Figure 5a shows monthly variations of SEP and sunspot number during the study

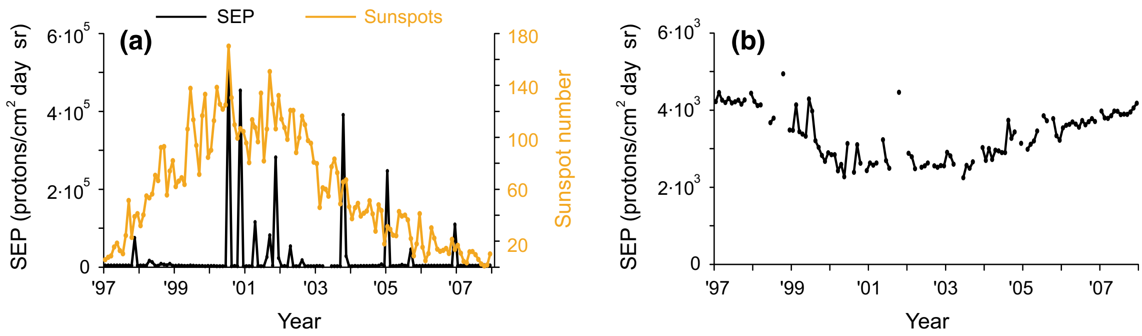

Fig. 5 **a** Time series of >100 MeV proton fluence and sunspot number. **b** Monthly variation of solar energetic protons without outliers

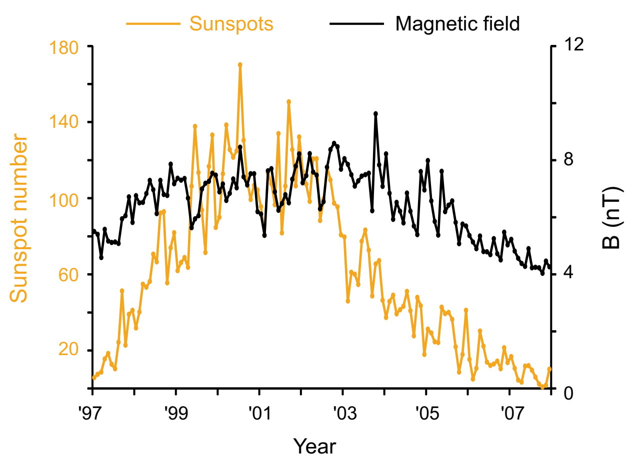

Fig. 6 Monthly variation of sunspots number and magnetic field magnitude

period. The largest pick events correspond to the maximum solar activity and the descending phase, i.e. the active periods of the solar cycle. When removing the outliers detected with Grubbs' test, the SEP time series (Fig. 5b) shows a similar behaviour than cosmic rays (Fig. 7a).

The larger solar flares can cause massive ejection of solar matter which are related with these outliers, and correspond mainly with optical flares of importance three, that is, optical flares which corrected area in heliospheric square degrees[1] at maximum brightness range from 12.5 to 24.7 square degrees (almost 60 % of this flares are related to these outliers). Correlation between >100 MeV proton fluence without outliers and ^{7}Be concentrations shows a weak significant positive correlation with a correlation coefficient $r = 0.30$. This correlation is weaker for lower energetic SEP, and it could be explained by the cut-off effects of the geomagnetic field, that only high-energy SEPs can reach the mid- and low-latitudes [33, 41].

[1] One square degree is equal to $(1.214 \times 104\ \text{km})^2 = 48.5$ millionths of the visible solar hemisphere.

Influence of cosmic rays in ^{7}Be variability

It is well known that interactions of cosmic rays with the terrestrial atmosphere produce radionuclides such as ^{7}Be, therefore there should be a positive correlation between ^{7}Be activity concentration and the flow of cosmic rays. Cosmic rays can be deflected by both the interplanetary magnetic field and solar wind which have an effect of shielding on the Earth. Solar activity and magnetic field magnitude have similar behaviour (Fig. 6) showing a strong positive correlation ($r = 0.69$). In turn, magnetic field magnitude and cosmic rays are anti-correlated with a correlation coefficient of $r = -0.77$. Thus, a lineal model to describe the relationship between both variables shows that the 59.97 % of the cosmic rays could be explained by fluctuations of the magnetic field. Consequently, when solar activity presents the maximum, cosmic ray amount reaching the Earth is the lowest (Fig. 7a), resulting in a reduced ^{7}Be production rates with increasing solar activity. This is in agreement with results of previous works [42–44].

The amount of ^{7}Be that reaches the Earth's surface is dependent on its atmospheric production rate, stratosphere–troposphere air exchange, and tropospheric circulation [45–47], and as a result, the flux of ^{7}Be from the atmosphere to Earth and cosmic rays show large scatter and rather low significant correlation ($r = 0.24$, p value $= 0.007$) (Fig. 7b). The correlation increases considerably ($r = 0.52$) when performing the correlation between the trends of ^{7}Be and cosmic rays. Then we can state that again meteorological factors may affect the relationship between both variables.

Influence of airborne particulate matter in ^{7}Be variability

As ^{7}Be becomes an aerosol associated species, a correlation between its specific activity and aerosol concentration is expected to be found. Aerosol optical depth (AOD) provides an estimation of sun light extinction due to atmospheric aerosols, and it is therefore an indirect

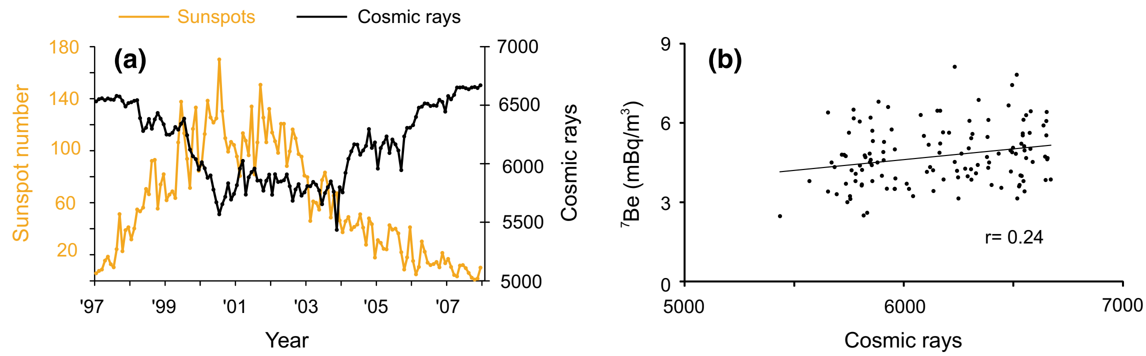

Fig. 7 **a** Comparison of monthly number of sunspots and flow of cosmic rays arriving at Earth for the period 1997–2007. **b** Monthly ^{7}Be activity concentration plotted against cosmic ray amount. The *solid line* represents the linear least squares' regression line

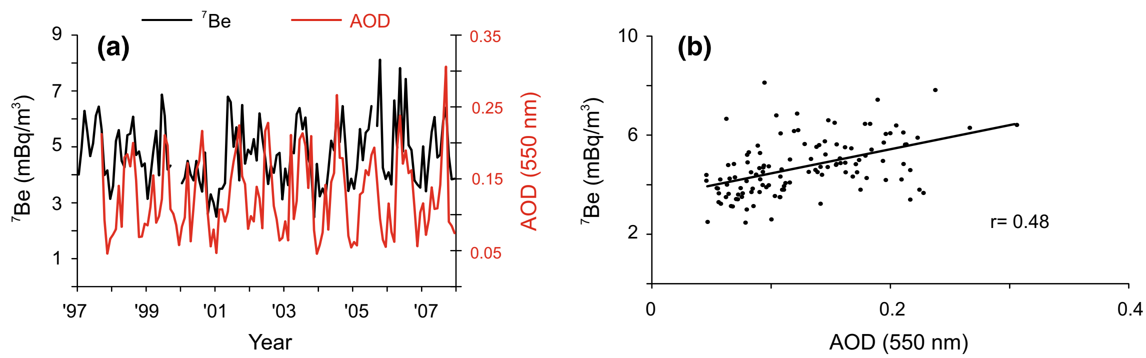

Fig. 8 **a** Monthly mean AOD_{550} and ^{7}Be specific activity for the period 1997–2007. **b** Scatter plot of monthly ^{7}Be atmospheric concentration versus monthly average SeaWiFS AOD_{550}. The linear regression line is shown as the *black line*

measurement of aerosols distributed within a column of air. We have used the AOD derived from Sea-viewing Wide Field-of-view Sensor (SeaWiFS) Deep Blue Level 3 satellite data with the retrieval algorithm over land and ocean [48, 49] at the reference wavelength of 550 nm and $0.5° \times 0.5°$ resolution to determine whether there is a significant correlation between aerosol concentration and ^{7}Be atmospheric concentration.

Figure 8a depicts the comparison between AOD and ^{7}Be activity concentrations. Both variables show a seasonal cycle with higher values in the spring and summer where differences do not reach statistical significance for ^{7}Be values according to the corrected Mann–Whitney test, and lower values in winter and autumn with non statistical differences in both variables. This pattern was also found by Dueñas et al. [7] and Cesnulyte et al. [50]. The higher AOD in spring–summer may be explained by several contributions such as stagnant synoptic meteorological patterns or secondary aerosol photochemical formation [51], as well as the high frequency of African dust outbreaks which occurs during these months in this area [52]. ^{7}Be spring–summer maximum could be attributed to the air exchange between the stratosphere and troposphere due to the thinning of the tropopause [6, 7] and the high occurrence of African dust outbreaks [7]. Figure 8b presents the performance of ^{7}Be specific activity versus AOD at 550 nm. There is a significant positive correlation with a correlation coefficient of $r = 0.48$, ratifying the attachment of ^{7}Be on airborne particles. When performing that correlation seasonally, it is found that only autumn does not show a statistically significant relationship. As AOD is an indirect measure of atmospheric particulate matter, and it depends on several factors like vertical structure, composition, size distribution and water content of atmospheric aerosol, further research is needed to assess the relationship between ^{7}Be and atmospheric particulate matter.

Multiple correlation analysis

A multiple linear regression model was developed to explain the variability of ^{7}Be activity concentrations at Málaga using all the cosmogenic factors studied above (sunspot number, cosmic rays and SEPs >100 MeV), as well as the presence of atmospheric particulate matter. When using the observed values, only the SEP removing the outliers and AOD are statistical significant predictors of the concentration of ^{7}Be measured at ground level in Málaga, explaining about 34 % of the variance. However, when the trend component of the ^{7}Be variability is studied, cosmic rays is the unique variable which does not become a significant predictor, explaining around 56 % of the variability of ^{7}Be trend component. Therefore, these results may indicate that cosmic rays have not strong influence on controlling the variability of ^{7}Be activity concentration at this coastal city. Furthermore, as the explained variance is greater when trend components are used, it could suggest that meteorological parameters may mask the influence of the cosmogenic factor on the ^{7}Be variations.

Conclusions

^{7}Be atmospheric concentrations were measured during an 11-year period in Málaga, Spain (4.49°W; 36.73°N) covering the 23rd solar cycle. Monthly ^{7}Be values range between 2.47 and 8.12 mBq m^{-3} with an average of 4.71 mBq m^{-3}. Concentrations of the cosmogenic radionuclide display seasonal trend with the highest values being observed in the spring–summer and the lowest in autumn–winter period.

The present study contributes to the understanding the relationship between the ^{7}Be and the variables solar activity (displayed by sunspot number and SEPs), cosmic ray and atmospheric particulate matter. During the active phase of the solar cycle, the sunspot number increases as well as the number of solar flare injections. A weak anti-correlation ($r = -0.14$) between ^{7}Be and sunspot number is found in good agreement with previous reported results. When removing the seasonal and the irregular component of the time series, the correlation increases ($r = -0.59$) suggesting that meteorological factors may mask the solar influence.

The intense solar flares can cause massive ejection of SEPs being outliers in the time series. If these outliers are eliminated, the SEP >100 MeV time series display a similar pattern than the one of the cosmic rays. Both variables show minimum values when maximum values of sunspot number (i.e. solar active phase) are found. There is a significant positive correlation between these variables and ^{7}Be ($r = 0.30$ and $r = 0.24$ for SEP and cosmic rays respectively), and hence an anti-correlation with sunspot number. This is due to the fact that the Sun's magnetic field is stronger during the solar maximum, and shields the Earth from cosmic rays and solar particles. However, the larger solar flares related mainly with optical flares of importance three, are not deflected by the magnetic field and can reach the Earth's atmosphere. Finally, a positive significant correlation is found between the AOD and ^{7}Be atmospheric concentrations ($r = 0.48$).

Acknowledgments This work has been supported by the Consejo de Seguridad Nuclear (CSN). We thank the Deep Blue science team for their efforts in producing the SeaWiFS Deep Blue aerosol data records. The authors gratefully acknowledge SPDF, NOAA, SWPC and the Sodankyla Geophysical Observatory of the University of Oulu for providing the data. We would also like to express our gratitude to Jose Antonio García Orza for lending us to use his computers for the simulations.

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