Analysis of the influence of solar activity and atmospheric factors on ^{7}Be air concentration by seasonal-trend decomposition

1. Introduction

^{7}Be is a cosmogenic radionuclide that has been widely studied in recent decades and used as an atmospheric tracer of aerosols due to its relatively short life ($T_{1/2} = 53.3$ days) and ease of measurement by γ-spectrometry. This radionuclide is formed by spallation reactions of light atmospheric nuclei (such as carbon, nitrogen and oxygen) with very high-energy protons and neutrons of the primary cosmic rays, with galactic cosmic rays the component mainly responsible for their production (Lal et al., 1958; Bruninx, 1961, Vogt et al., 1990; Baeza et al., 1996). Most ^{7}Be production (~70%) occurs in the stratosphere and the remainder (~30%) is produced in the troposphere, consequently the ^{7}Be production rate is altitude-dependent (Feely et al., 1989).

The intensity of galactic cosmic rays in the Earth's orbit is affected by solar activity and the geomagnetic field, under constant cosmic ray bombardment in space (Vogt et al., 1990). Solar modulation of galactic cosmic rays shows a cyclical trend of approximately 11 years and previous studies have confirmed the inverse correlation with solar activity as a result of the shielding caused by the magnetic activity of the Sun (O'Brien, 1979; Hötzl et al., 1991; Ioannidou and Papastefanou, 1994). Solar activity has a direct relationship with the sunspot number and for this reason, this parameter is widely used to represent solar activity (Cannizzaro et al., 1995; Papastefanou and Ioannidou, 2004; Piñero-García and Ferro-García, 2013). Kotsopoulou and Ioannidou (2012) pointed out that solar activity is the dominant cause of the observed variation of galactic cosmic rays. Moreover, the geomagnetic field deflects incoming galactic cosmic rays toward the magnetic poles, so that the ^{7}Be production rate is much higher at high latitudes (Feely et al., 1989).

In addition to the above-mentioned sources of variability, ^{7}Be concentration in the lower layers of the atmosphere presents temporal variations caused by other factors. Thus, the concentration of this radionuclide has a seasonal pattern with maximum values in spring-summer and minimum values in autumn-winter. This seasonality is mainly explained by the following factors (Feely et al., 1989; Baeza et al., 1996):

i) the incorporation of ^{7}Be into the lower layers of the troposphere due to the displacement of masses of air from higher layers caused by solar radiation. According to Baeza et al. (1996), the maximum values of ^{7}Be concentrations in the hottest months may be justified by vertical air mixing. The rising hot air forces the cold air in the higher layers to move downwards, where there are greater concentrations of cosmogenic radionuclides. The heating of air masses is directly related to the longer length of the solar day and the greater angle of incidence of solar radiation during the summer.
ii) the meteorological parameters that affect the climatology of regions in which the sites are located, such as temperature, relative humidity, precipitations, and wind speed.

The aim of this paper is to study the different sources of variability of ^{7}Be air concentration in the city of Valencia over the period 2007-2014, using a methodology that decomposes a time series into different components. The decomposition model makes it possible to estimate the influence of solar activity and atmospheric factors on the independent components, in order to find the different sources of ^{7}Be variability. It should be noted that we focus on the ^{7}Be concentration in the air of the city of Valencia, which has a fixed latitude and altitude, so that the variability caused by these factors is not considered.

2. Material and methods

2.1. Sampling

Airborne particulate samples were collected weekly on the campus of the Universitat Politècnica de Valencia from January 2007 to December 2014. Valencia is situated on the east coast of Spain (15m above sea level) in the western Mediterranean Basin (39°28′50″ N, 0°21′59″ W) (Fig. 1) and has a relatively dry subtropical Mediterranean climate with very mild winters and long hot summers. The sampling point was located approximately 2 km away from the coastline, so that high relative humidity is registered, especially in summer.

Aerosol samples were collected using Eberlyne G21DX and Saic AVS28A air samplers that were situated approximately 1 m above ground. The aerosol particles were retained on a cellulose filter of 4.2 x10^{-2} m effective diameter and 0.8 µm pore size. Filters were changed weekly and the average volume ranged from 300 to 400 m^3 per week. Each filter was introduced in a plastic box and kept in a desiccator until its measure.

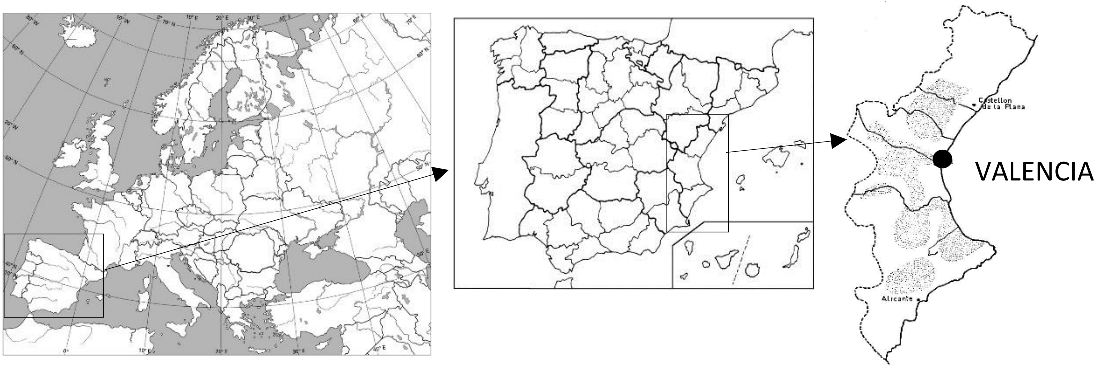

Fig. 1. Map showing the location of Valencia in the center-eastern coast of the Iberian Peninsula

2.2. *^{7}Be activity measurements*

A monthly composite sample containing 4-5 filters was measured by γ-spectrometry to determine specific ^{7}Be activities using an n-type HPGe detector (ORTEC Industries, USA) with a relative efficiency of 18% for 60Co gamma-rays. A certificated standard containing radionuclides with energies ranging from 59 to 1836.1 keV was used for preparing calibrated filters. The filters were placed on the top of the detector inside a plastic box. The counting time was 60000s and the γ-line 477.7 KeV was used to calculate the activity. ORTEC Gamma-Vision software was used for acquisition and analysis. Concentration activities were corrected for the radioactive decay to the mid-collection period. The mean measured uncertainties (K=2) were around 10 %.

2.3. *Solar activity and atmospheric factors*

Solar activity as measured by the sunspot number parameter was considered. A sunspot is a region on the Sun with a strong magnetic field and lower temperatures than the surrounding photosphere. The sunspot number parameter (SSN) was collected daily during the period 2007-2014 by the World Data Center SILSO, Royal Observatory of Belgium, located in Brussels (SILSO, 2015).

The meteorological factors studied in the present work are: precipitations (PP) (in tenths of millimeters), temperature (T) (in tenths of °C), relative humidity (RH) (%) and wind speed (W) (km/h), collected by the meteorological station at the Universitat Politècnica de Valencia, and solar radiation (KJ/m^2) provided by the Spanish National Institute of Meteorology, AEMET.

In this study we considered the mean monthly values of temperature, relative humidity, wind speed, sunspot number and direct solar radiation. The precipitation factor was considered as the number of rainy days per month due to the particular rainfall regime in Valencia, with few days of torrential rainfall and many dry days.

2.4. *Statistical methods*

2.4.1. *Exploratory analysis*

A descriptive analysis was applied to study the behavior of ^{7}Be activity during the period 2007-2014. First, the distribution of this activity was characterized and the main descriptive statistics were obtained to analyze the data. The analysis of variance test (ANOVA) was then carried out to study the statistical significance of the differences in ^{7}Be concentration in the air over the years and seasons.

2.4.2. Seasonal-trend decomposition of a time series

The variation of ^{7}Be data with time was studied by time series analyses. A time series is a sequence of observations taken sequentially in time. Many procedures can be used to analyze a time series. We opted to decompose the ^{7}Be time series into a mixture of four independent components (Kendall, 1976):

i) a *trend component* (T_t) or long-term movement, that reflects the long term variation of the time series.
ii) a *cycle component* (C_t) or fluctuations about the trend of greater or less regularity.
iii) a *seasonal component* (S_t), reflecting seasonality. Seasonal variations are short-term fluctuations in a time series which occur periodically in a period of time (in this study the period of time is one year).
iv) an *irregular component* (I_t), or residual effect that describe random and irregular influences.

It is interesting to represent series as the sum of these four components and one of the objects of the analysis is to break the series down into their constituents for individual study.

Let Y_t , $t = 1, \dots, N$ denote a time series from which the unobserved trend component has to be extracted. It is very difficult to distinguish between the trend and the cycle components (this requires long series and complete cycle data being available), so in general both components are combined into one named trend-cycle (TC_t). In this study we also considered the trend-cycle component in the time series, since the study period is short (2007-2014):

$$Y_t = TC_t + S_t + I_t \qquad (1)$$

There are many techniques for depicting the trend-cycle component from a time series using filters or parametric regression models. The most common forecasting technique to estimate the trend-cycle component, which does not require any assumption, is the symmetric centered moving average filter of order s defined by the following expression (Kendall, 1976):

$$\widehat{TC}_t = \frac{1}{s} \sum_{k=-s/2}^{s/2} w * Y_{t+k}, \qquad (2)$$

$$w = 0.5 \; for \; k = \{-s/2, s/2\}; \; w = 1 \; \forall k \neq \; \{-s/2, s/2\}; \; (s/2 + 1) \leq t \leq N - s$$

In the present study $s = 12$, because the objective is to study the trend-cycle component over a number of months of a year (inter-annual variability). Once the trend-cycle component has been estimated, the times series:

$$Y'_t = Y_t - \widehat{TC}_t = S_t + I_t \qquad (3)$$

is called a *detrended* series. It should be noted that Y'_t does not start to yield values until $t = 7$.

In order to estimate the seasonal factors $\widehat{S}_t$ of the *detrended* series Y'_t, we used a stable seasonal filter. For observations made during period $k, k = 1, .., s$ a stable seasonal filter computes a single seasonal value for each month by first calculating the average of all the values for each month (taken after the detrending adjustment), which reduce the change of the irregular component affecting the seasonal component (Kendall, 1976):

$$\widetilde{S_k} = \frac{1}{(N/s)} \sum_{j=1}^{(N/s)} Y'_{k+js}\,, \quad k = 1, \ldots, 6$$

$$= \frac{1}{(N/s)} \sum_{j=0}^{(N/s)-1} Y'_{k+js}\,, \quad k = 7, \ldots, 12 \qquad (4)$$

The seasonal factors are defined as:

$$\widehat{S_t} = \widetilde{S_k} - \bar{S}, \qquad \bar{S} = \frac{1}{s}\sum_{i=1}^{s} \widetilde{S_k}\,, \qquad t = 1, \ldots, 12 \qquad (5)$$

In Eq. (5) we impose the condition that $\sum_{i=1}^{s} \widehat{S_t} = 0$ to define the seasonal factor fluctuation around zero. Given estimates $\widehat{TC_t}$ and $\widehat{S_t}$, the irregular component is estimated as:

$$\widehat{I_t} = Y_t - \widehat{TC_t} - \widehat{S_t} \qquad (6)$$

The above-described method was found to be useful in the present study to estimate the influence of the different factors (solar activity and atmospheric parameters) on the trend-cycle and on seasonal and irregular components of the ^{7}Be time series.

3. Results and discussion

3.1. Temporal evolution of ^{7}Be air concentration

The temporal evolution of ^{7}Be air concentration at the Universitat Politècnica de València from 2007 to 2014 is shown in Fig. 2. Goodness of fit to normality was proved by the Kolmogorov-Smirnov test. As the p-value obtained was 0.57, the monthly ^{7}Be air concentration fits well with a normal distribution. The arithmetic mean was therefore used as a descriptive statistic to analyze the data.

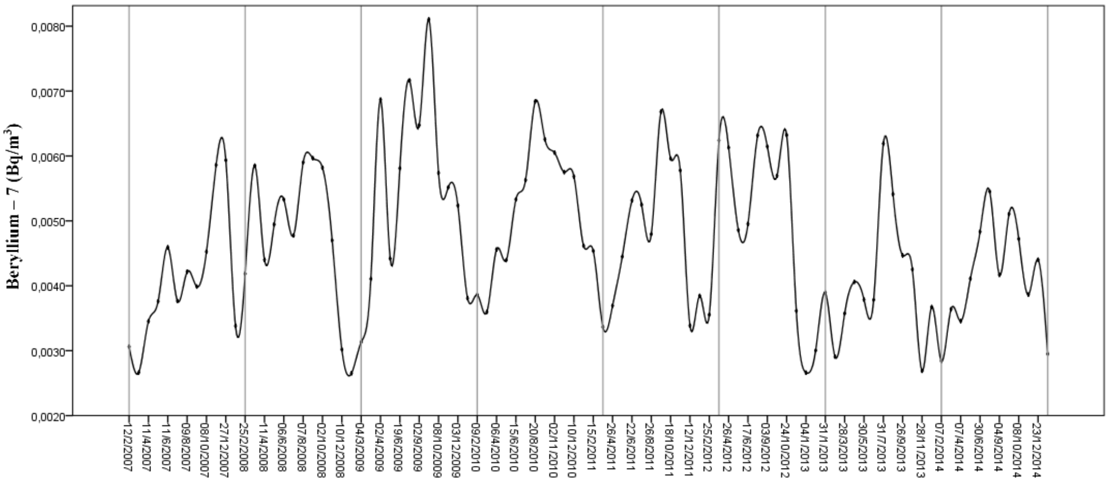

Fig. 2. Temporal evolution of ^{7}Be air concentration at Universitat Politècnica de València over the period 2007-2014.

^{7}Be activity concentrations ranged from 2.65 to 8.11 mBq/m^3 with an arithmetic mean of 4.69±1.20 mBq/m^3 during the period studied. The values obtained are consistent with ^{7}Be air concentrations reported in previous studies in regions at similar latitudes, such as Granada (5 mBq/m^3) (Azahra et al., 2004a), Málaga (4.17 mBq/m^3) (Piñero-García and Ferro-García, 2013), Cáceres (4.4 mBq/m^3) (Baeza et al., 1996), Palermo (5.06 mBq/m^3) (Cannizzaro et al., 2003).

The concentration of ^{7}Be in the monthly samples collected and analyzed from 2007 to 2014 are reported per year and season. Table 1 lists the arithmetic mean of the annual ^{7}Be activity concentrations and the corresponding standard deviation. The highest mean concentration is observed in 2009, coinciding with the minimum solar activity of the 24th solar cycle. The result obtained is in agreement with the findings of Piñero-García and Ferro-García (2013). On the other hand, the lowest mean concentration occurred in 2013, which coincided with the maximum of the 24th solar cycle.

Table 1. Annual ^{7}Be activity concentrations in surface air of Valencia

Year	^{7}Be (mBq/m^{3})
2007	4.10±1.01
2008	4.80±1.10
2009	5.53±1.48
2010	5.21±1.00
2011	4.75±1.07
2012	4.96±1.40
2013	4.06±0.97
2014	4.13±0.82

As can be seen in Table 2, the maximum ^{7}Be concentration was observed during the spring-summer months and the minimum during the autumn-winter months.

Table 2. Seasonal ^{7}Be activity concentrations in surface air of Valencia

Season	^{7}Be (mBq/m^{3})
Winter	4.07±1.14
Spring	4.88±0.85
Summer	5.65±0.93
Autumn	4.18±1.14

The ANOVA test was applied to study the significance of the differences in ^{7}Be concentrations between annual and seasonal factors. The p-values obtained are less than 0.05 for each factor ($p - value < 0.00001$, $p - value = 0.0005$, for annual and seasonal factors respectively), which means the differences in ^{7}Be activity concentrations are statistically significant at a 95% confidence level. Fig. 3 shows the arithmetic mean for each factor level and the Bonferroni intervals used to observe significant differences. Fig. 3 (a) shows the inter-annual variability and Fig. 3 (b) shows the intra-annual variability of ^{7}Be air concentrations, which were as expected.

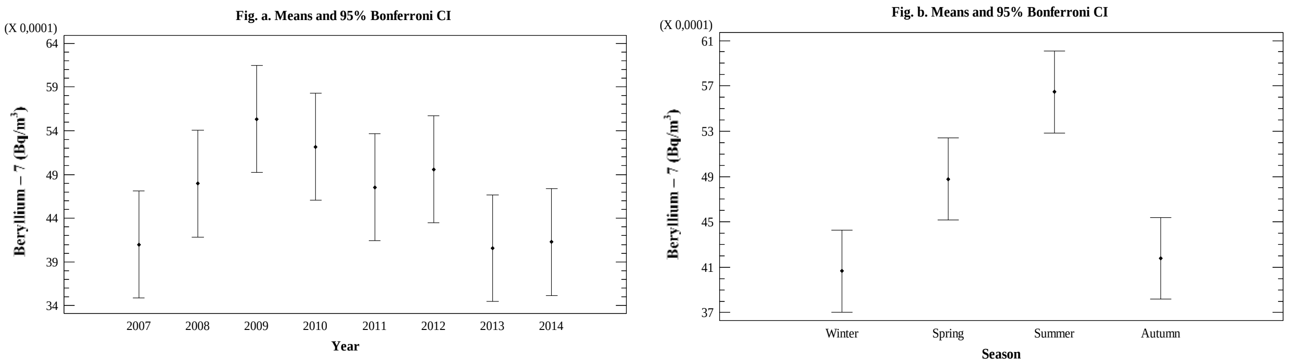

Fig. 3. Means and 95% Bonferroni CI for the effect of (a) annual and (b) seasonal factors in ^{7}Be activity concentration

3.2. *Seasonal-trend decomposition into ^{7}Be time series*

Following the method described in Section 2.4.2, the ^{7}Be time series is decomposed into a sum of three independent components (trend-cycle, seasonal and irregular components):

$$ {}^{7}Be_t = \widehat{TC}_t + \widehat{S}_t + \widehat{I}_t \qquad (7) $$

Fig. 4 shows the trend-cycle component, $\widehat{TC}_t$, which explains the inter-annual variability of the ^{7}Be air concentration as expected. Fig. 5 shows the seasonal component of the ^{7}Be row data, $\widehat{S}_t$. Note that the behavior of the seasonal component is regular throughout these years, so that this component explains intra-annual ^{7}Be variability. However, the ^{7}Be data is not constant throughout the years, as there is an irregular component, $\widehat{I}_t$, (Fig. 6), which explains the variability observed due to the residual effect that describes random and irregular influences.

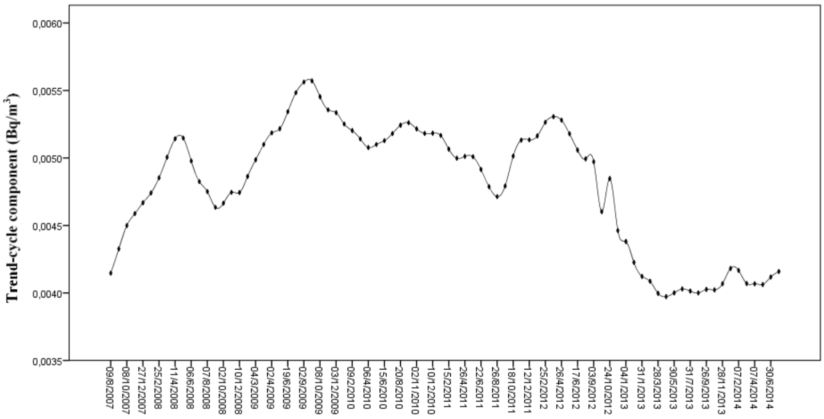

Fig. 4. Trend-cycle component obtained by time series decomposition over the period 2007-2014.

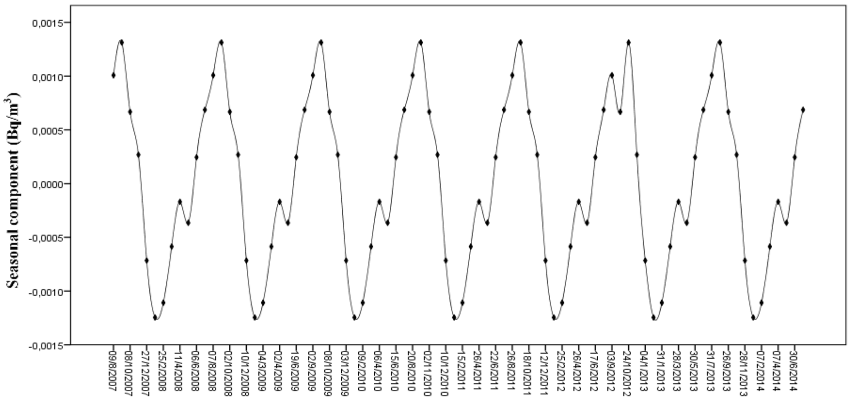

Fig. 5. Seasonal component obtained by time series decomposition over the period 2007-2014.

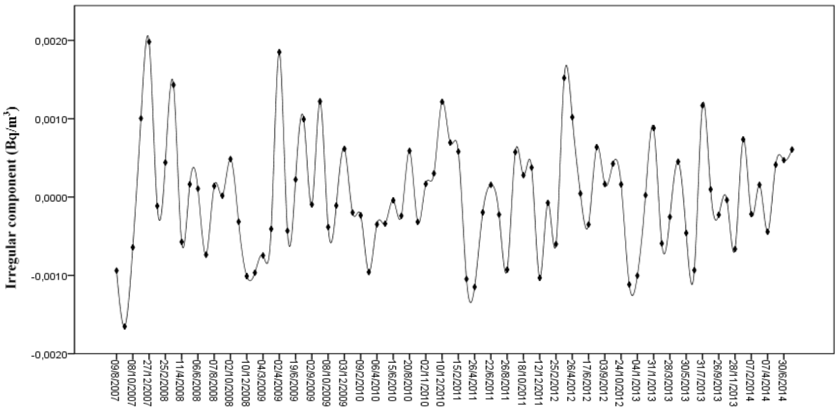

Fig. 6. Irregular component obtained by time series decomposition over the period 2007-2014.

Multiple regression was used to quantify the actual relative importance of each component in ^{7}Be variability, obtaining the standardized coefficients of each component (Saltelli et al., 2008). The results show that the trend-cycle component explains 23.60%, the seasonal component 36.28% and the irregular component 40.12% of ^{7}Be variability. The seasonal and irregular components have a strong influence on ^{7}Be variability, which shows it is important to study its dependence on atmospheric parameters in order to find sources of variability that explain ^{7}Be activity.

3.3. Influence of solar activity and atmospheric parameters in ^{7}Be row data, $\widehat{TC_t}$, $\widehat{S_t}$ and $\widehat{I_t}$ components

Pearson's linear correlations between ^{7}Be row data and the time series components with sunspot number and atmospheric parameters are given in Table 3.

Table 3. Pearson's correlations between trend-cycle, seasonal and irregular components and the solar activity and atmospheric factors. The p-values are reported in brackets.

	Description	SSN	SR	T	PP	W	RH
^{7}Be	Row data	-0.2140 **(0.0363)***	0.5029 **(<0.00001)****	0.5801 **(<0.00001)****	-0.2484 **(0.0147)***	-0.2947 **(0.0036)****	0.4374 **(<0.00001)****
$\widehat{TC_t}$	Trend-Cycle component	-0.5045 **(<0.00001)****	-0.0223 (0.8405)	-0.0541 (0.6253)	0.1181 (0.2847)	-0.1688 (0.1247)	0.0514 (0.6421)
$\widehat{S_t}$	Seasonal component	-0.0571 (0.6061)	0.6606 **(<0.00001)****	0.9441 **(<0.00001)****	-0.2334 **(0.0326)***	-0.1700 (0.1221)	0.6027 **(<0.00001)****
$\widehat{I_t}$	Irregular component	-0.0437 (0.6934)	0.2298 **(0.0355)***	0.0017 (0.9874)	-0.2724 **(0.0122)***	-0.2254 **(0.0392)***	-0.0388 (0.7261)

** significant correlations at 95% confidence level*

*** significant correlations at 99% confidence level*

The trend-cycle component ($\widehat{TC_t}$) has a strong negative correlation with the SSN parameter ($r = -0.50$) and a non-significant correlation with the atmospheric variables. However, the correlation between ^{7}Be row data and SSN is rather weak ($r = -0.21, p - value = 0.036$). This shows that the atmospheric variables may mask the solar influence. Recent studies have also reached the same conclusion (Dueñas et al., 2015). As the seasonal and irregular components are not correlated with the SSN parameter, the atmospheric variables may be the source of the variability of the seasonal and irregular components.

From Figure 7 it is evident that the trend-cycle component and the sunspot number parameter are strongly and inversely correlated.

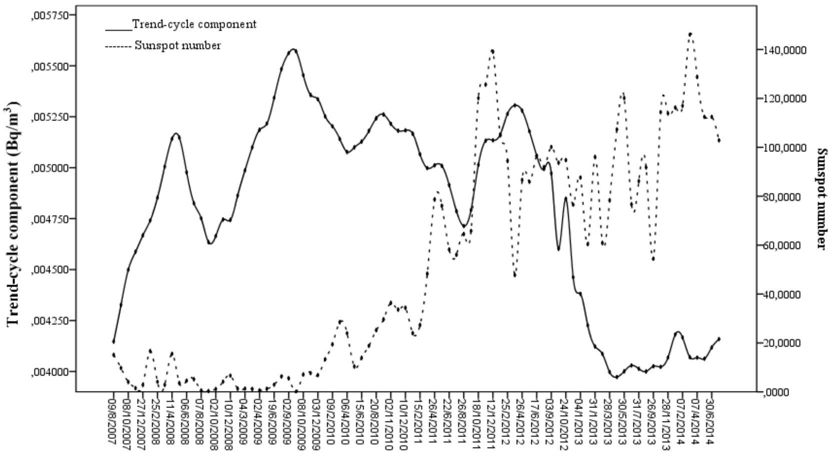

Fig. 7. Temporal evolution of sunspot number and trend-cycle component over the period 2007-2014

The seasonal component ($\widehat{S_t}$) has a strong positive correlation with solar radiation, temperature and relative humidity. However, it has a weak negative correlation with precipitations and it is not correlated with wind. Neither are solar radiation, temperature and relative humidity correlated with the trend-cycle component, but they are weakly correlated with the irregular component.

Figures 8, 9 and 10 show that solar radiation, temperature and relative humidity have a regular trend over the years similar to that of the seasonal component. In general, these atmospheric variables are higher in spring-summer than in autumn-winter periods.

^{7}Be row data has also a positive correlation with solar radiation, temperature and relative humidity; however, this correlation is strengthened with the seasonal component. The correlation observed between the seasonal component and solar radiation is positive ($r = 0.66$), and is higher than the correlation observed with ^{7}Be row data ($r = 0.50$). The seasonal component is strongly and positively correlated with the air temperature ($r = 0.94$). This correlation is much stronger than the correlation observed with ^{7}Be row data ($r = 0.58$). Thus, the seasonal-trend decomposition reveals the strong influence of temperature on the seasonal variability of the ^{7}Be air concentration.

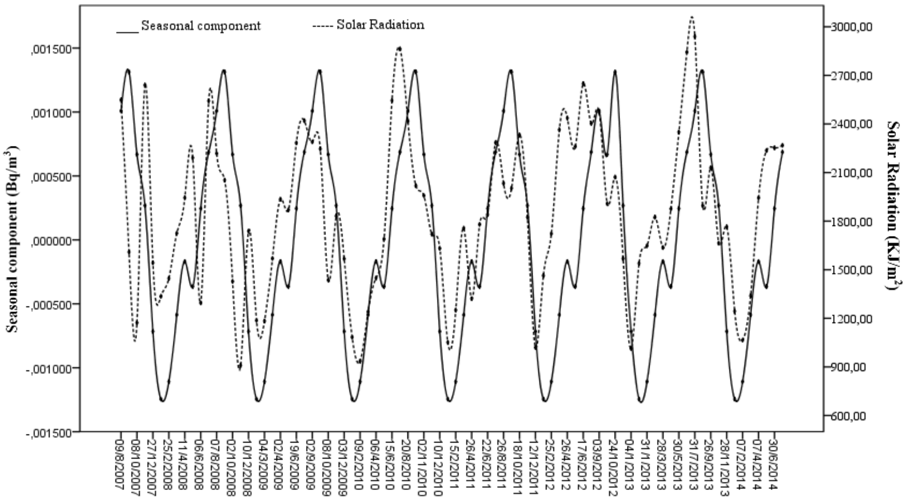

Fig 8. Temporal evolution of solar radiation and seasonal component over the period 2007-2014.

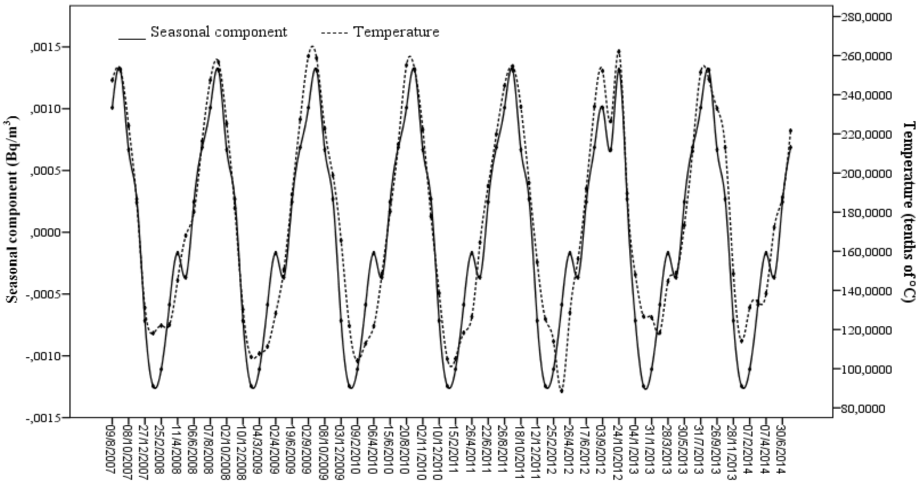

Fig 9. Temporal evolution of temperature and seasonal component over the period 2007-2014.

Many studies have observed a negative correlation between ^{7}Be air concentration and relative humidity (Dueñas et al., 2009; Kotsopoulou and Ioannidou, 2012; Piñero-García and Ferro-García, 2012). However, in this study RH is positively correlated with the seasonal component ($r = 0.60$). This correlation is higher than the correlation observed with ^{7}Be row data ($r = 0.43$) (Fig. 10). The positive relationship may be ascribed to the particular meteorological conditions in Valencia, which has characteristically high relative humidity, especially in summer. This would seem to indicate that there are contrary relations in term of different RH. With the same RH, the aerosol residence time can differ due to other meteorological conditions that would result in different removal of ^{7}Be from surface air and influencing its contents in the air, which would thus be different to previous studies.

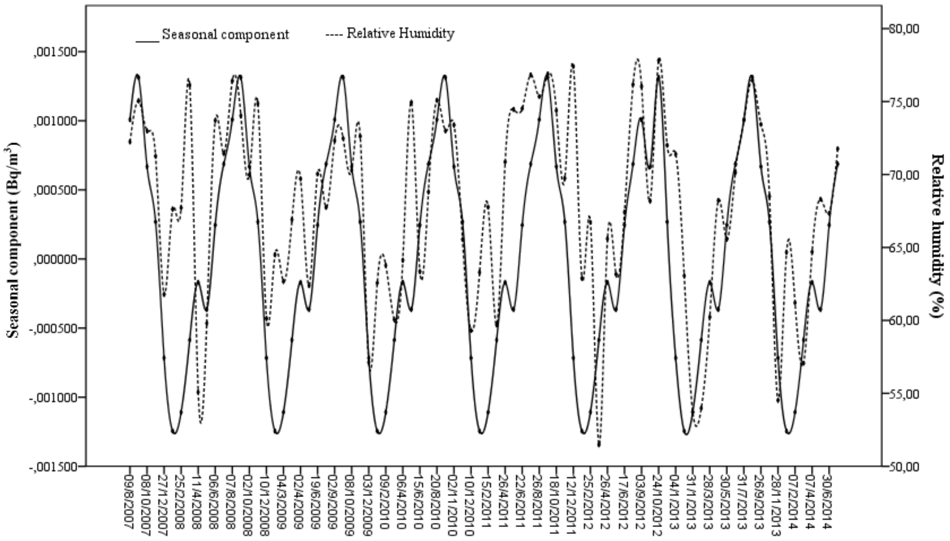

Fig 10. Temporal evolution of relative humidity and seasonal component over the period 2007-2014.

The irregular component ($\widehat{I_t}$) is negatively correlated with precipitation ($r = -0.27$) and wind speed ($r = -0.23$) and these negative relationships are also observed with ^{7}Be row data. Note that precipitations and wind speed are not correlated, or are weakly correlated, with the seasonal and trend-cycle components. Thus, precipitations and wind speed are influential factors on the irregular variability of ^{7}Be air concentration.

The correlation observed with the precipitations is supported by recent studies, which found a strong correlation between precipitations and ^{7}Be deposition fluxes (Chen et al., 2016; Zhang et al., 2016). In constrast, many studies have found a non-significant correlation between ^{7}Be row data and the total monthly precipitation per month (Azahra et al., 2004b; Ali et al., 2011; Pham et al., 2011; Kotsopoulou and Ioannidou, 2012).

Other studies, e.g. Dueñas et al. (1999, 2009), Lozano et al. (2012), Kusmierczyk-Michulec et al. (2015), found little or no evidence of an inverse relationship between wind speed and ^{7}Be concentration. However, in this study, the relationship found between wind speed, the ^{7}Be row data and the irregular component is significant and negative.

Figures 11 and 12 show the relationship between precipitations and wind speed, and the irregular component, indicating that this radionuclide is washed out of the air by rain and wind, which reduces ^{7}Be residence time in the air.

The irregular component has a non-significant or weak correlation with temperature, solar radiation and relative humidity, which are atmospheric factors with a regular pattern over the years and highly correlated with the seasonal component.

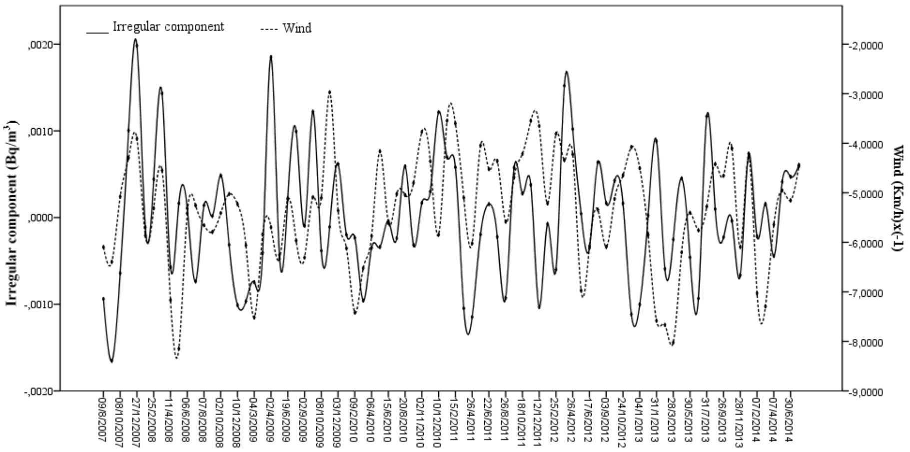

Fig 11. Temporal evolution of wind speed factor and irregular component over the period 2007-2014. Note that the negative values of wind speed are represented to better identify the relationship.

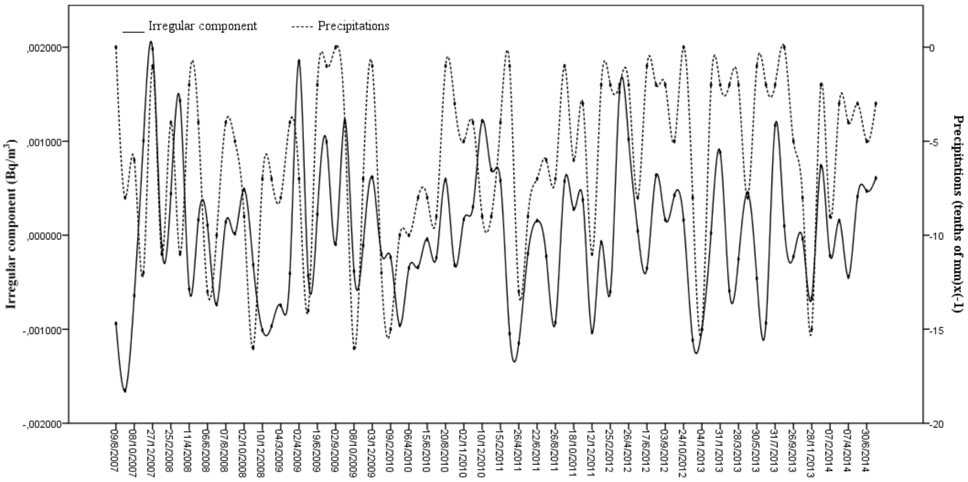

Fig 12. Temporal evolution of precipitation and irregular component over the period 2007-2014. Note that the negative values of precipitation are represented to better identify the relationship.

4. Conclusions

^{7}Be air concentrations were measured in Valencia, Spain, over the period 2007-2014. The mean values of monthly ^{7}Be concentrations ranged from 2.65 to 8.11 mBq/m^3, with an average of 4.69±1.20 mBq/m^3. ANOVA analysis confirmed that the intra and inter-annual variability of the ^{7}Be activity was as expected.

The use of seasonal-trend decomposition has been shown to be a powerful method of describing the ^{7}Be time series as a sum of independent components responsible for different patterns of ^{7}Be variability.

Solar activity is a cosmogenic factor with a high influence on the trend-cycle component of ^{7}Be variability, which increases when solar activity is minimum. This inverse relationship is clearer after removing seasonal and irregular components of the ^{7}Be time series, suggesting that atmospheric factors may mask the solar influence.

Solar radiation, temperature and relative humidity are influential factors in seasonal ^{7}Be variations. ^{7}Be concentrations increase with higher solar radiation and air temperature. The seasonal-trend decomposition revealed the particularly strong influence of temperature on seasonal variability. Relative humidity is also positively correlated with ^{7}Be concentrations. This result disagrees with previous studies, and may be due to the particular meteorological conditions in Valencia, with high relative humidity, especially in summer.

A negative relationship was found between ^{7}Be row data and precipitations and wind speed, even though many previous studies found little or no evidence of this relationship. Seasonal-trend decomposition allowed us to identify the negative relationship of these atmospheric factors with the irregular component. It can therefore be said that precipitations and wind speed are influential factors in the irregular variability of ^{7}Be air concentration.

These results thus show the advantages of using seasonal-trend decomposition to separate the different patterns of ^{7}Be variability as a preliminary step in developing a predictive model based on time series analysis.

5. Acknowledgements

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