Gross alpha, gross beta activities and $^7$Be concentrations in surface air: analysis of their variations and prediction model

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Abstract

Measurements of gross $\alpha$, gross $\beta$ activities and cosmogenic beryllium-7 ($^7$Be) concentrations were made each week during the years 1992, 1993, 1994 and 1995 from samples of atmospheric aerosols filtered from the air. The data are sufficiently numerous to allow us to examine variations in time and through these measurements we have established several parameters that should be of importance in understanding any trends in radionuclides concentrations in the atmosphere. The weekly variation is discussed in relation to various meteorological factors. Seasonal variation of $^7$Be concentration in the atmosphere was measured and there was a tendency for a maximum in the spring and summer and a minimum in fall and winter. It can be said for this radionuclide that our results showed a prevailing influence of weekly average of the daily maximum temperature. In this work, the data on concentrations and meteorological data have been made use of in order to determine a model for gross $\alpha$, gross $\beta$ and $^7$Be, respectively. The model can be used to estimate that part of the trend in gross $\alpha$ and gross $\beta$ activities and in $^7$Be levels that can be accounted for by trends in local meteorology. A satisfactory agreement between the results of the model and the measurements was highlighted. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Gross $\alpha$ activity; Gross $\beta$ activity; $^7$Be concentrations

1. Introduction

Radioactivity in the atmosphere originates from natural radioactive decay, cosmogenic production and from nuclear weapon testing and nuclear accidents. Inhalation is one of the pathways by which radioactive nuclides are incorporated into the human body. Atmospheric radionuclide monitoring is only a means to make decisions and take provisions for mitigating the risk of disease reaching the general public, should there be an emergency. Emergency of a nuclear installation is a matter of relatively short duration. It is not the purpose of this paper to predict the impact of specific incremental radiation exposure arising from human activities of populations but with the information base provided, national and international bodies may be able to select appropriate criteria for the radiological protection of natural populations communities and ecosystems. In our laboratory, we are continuously measuring the radioactivity in the surroundings as part of a general project undertaken by many laboratories in Spain in collaboration with the Spanish authorities. In this project gross $\alpha$ and gross $\beta$ activities are routinely measured in samples of airborne dust samples and our sampling site is geographically far from the influence of nuclear installations.

Beryllium as well as other natural radionuclides participates in the formation and growth of the accumulation mode of aerosols (0.07 to 2 $\mu$m diameter) which is a major reservoir of pollutants in the atmosphere. $^7$Be is a relatively short-lived ($t_{1/2} = 53.3$ days) radionuclide of cosmogenic origin which is formed by spallation reactions, disintegration of nuclei of nitrogen and oxygen atoms that have been hit by cosmic ray neutrons (Bhandari et al., 1966). Once $^7$Be is formed in the troposphere it
rapidly associates primarily with submicron-sized aerosol particles. $^7$Be in these fine aerosols may subsequently enter the marine as well as the terrestrial and vegetation environment via wet or dry depositional events. Before deposition, $^7$Be will tend to associate with particulate material (Papastefanou and Ioannidou, 1994).

Several investigators (Young and Silker, 1974, 1980; Bleichrodt, 1978; Dutkiewicz and Husain, 1979; Dibb and Jaffrezo, 1998) have suggested that $^7$Be might prove useful in the study of atmospheric mixing and circulation. The high specific activity resulting from its half-life and the lack of dilution by stable beryllium make it an attractive choice for such work. Variations in concentrations of $^7$Be have often been used as an indicator of parcels of stratospheric air that have entered the troposphere (Gustafson et al., 1961; Viezee and Singh, 1980) but concentrations of $^7$Be at any particular point in the troposphere may also vary in response to other meteorological factors.

In this paper we report three years of continuous measurements of gross $\alpha$ and gross $\beta$ activities and concentrations of $^7$Be in surface air. Using these data, the present research was undertaken with the following principal goals:

1. To perceive the variations of gross $\alpha$ and gross $\beta$ and $^7$Be concentrations in Málaga.
2. To identify the main meteorological parameters that are responsible for the variations of those concentrations.
3. To model the data obtained from our sampling site as a function of meteorological parameters such as temperature, precipitation, wind speed, relative humidity, etc. With the regression model we believe that we can estimate the ground level concentrations associated with changes in meteorological conditions.

2. Experimental procedures

Airborne dust samples were collected weekly in cellulose nitrate filters, 47 mm diameter (collection efficiency 99.99% for 0.8 $\mu$m pore size) with an air sampler (RADECO, mod. AVS-28A) at a flow rate of 30 l min$^{-1}$, covering a total period from January 1992 to June 1995. Sample air volumes were determined by an in-line dry gas meter and about 300 m$^3$ ambient air per week have been drawn through the filters for the routine sampling that has been performed. Less than 3 mg of particulate material was collected in each experiment, thus overloading was excluded. The air sampler was lodged in an all weather sampling station and situated 10 m above the ground, on the roof of the Faculty of Science building, University of Málaga (4°28′8″W; 36°43′40″N) (see Fig. 1). Málaga is the capital of the province of the same name and it is in the southeast zone of the Iberian Peninsula on the Mediterranean coast. The climate in Málaga is warm, temperate with hot summers and little rain (550 mm yr$^{-1}$).

The instrumentation to count the gross $\alpha$ and gross $\beta$ activities was S$\alpha$n(Ag) and an $\alpha/\beta$ counter of the low-background multiple detector type (CANBERRA HT-1000), respectively. Air filters were kept in a desiccator for five days and counted in order to ensure complete decay

Fig. 1. Location of sampling point.
of daughter products of $^{222}$Rn. Since the levels of radioactivity encountered in environmental samples are low, long counting times were necessary, on the order of 1000 min sample$^{-1}$. The background of each detector was determined before and after use by 1000 min count. All the calculations have been made using the appropriate density thickness corrections for efficiencies to convert the gross-$\alpha$ (based on $^{241}$Am) and gross-$\beta$ (based on $^{90}$Sr) measurements to specific activities in Bq m$^{-3}$ with estimates of error at $\pm 1.96\sigma$.

Measurement of $^7$Be in each sample was carried out by non-destructive $\gamma$-ray spectrometry by means of its 477.6 keV $\gamma$-ray using a hyper-pure Ge-detector made by CANBERRA (relative efficiency about 30% to the efficiency of a $3`` \times 3``$ NaI(Tl) at 25 cm distance; resolution 2 keV for 1.33 MeV $\gamma$-ray of $^{60}$Co) connected to a 8192-channel pulse-height analyzer. The counting efficiencies of the Ge-detector were measured by using a standard sample containing known amounts of radioisotopes such as $^{133}$Ba, $^{152}$Eu, $^{137}$Cs and $^{60}$Co. Counting time for each sample was generally $9 \times 10^4$ s, leading to the detection limit below about 0.3 Bq sample$^{-1}$. The $^7$Be concentration was calculated using a 53 day half-life, $\gamma$ counting efficiencies of 3.2% and $\gamma$ branching ratios of 0.11. The concentrations were corrected for decay to the middle collection period.

3. Results and discussions

The gross-$\alpha$ and gross $\beta$ activities and $^7$Be data used in the analysis are weekly values of concentration in surface air. Aerosol sampling for atmospheric radionuclides commenced in January 1992 and terminated in July 1995 (177 data). Measurements of $^7$Be in each sample were carried out from January 1993 to June 1995 (122 data).

The results from individual measurements of the gross $\alpha$ and gross $\beta$ activities and $^7$Be concentrations were analyzed to derive the statistical estimates characterizing the distributions. Table 1 provides arithmetic mean (AM) and related statistical information such as geometric mean (GM), standard deviation (S.D.), dispersion factor of the geometric mean (DF), maximum and minimum values. These values are given ($\times 10^{-4}$) Bq m$^{-3}$ for gross $\alpha$ and gross $\beta$ activities and ($\times 10^{-3}$) Bq m$^{-3}$ for $^7$Be.

Plots of the frequency distribution show highly skewed (flat on the right) histograms for $\alpha$ and $\beta$ and a symmetric one for $^7$Be (Figs. 2–4). These trends have been quantified further using a graphical method (normal probability plot) and $\chi^2$ goodness-of-fit test. Log-normal distribution for $\alpha$ and $\beta$ data are significant at the 0.1–0.05 level. Otherwise, $^7$Be concentrations appear approximately normal. Assuming these types of distribution, the GM for the $\alpha$ and $\beta$ data and AM for $^7$Be data should be used to characterize average values (Sachs, 1984).

A range of values of $1.87 \times 10^{-4}$ Bq m$^{-3}$ and a geometric mean of $0.68 \times 10^{-4}$ Bq m$^{-3}$ were found for gross-$\alpha$ activity and a range of values of $12.18 \times 10^{-4}$ Bq m$^{-3}$ with a geometric mean of $5.20 \times 10^{-4}$ Bq m$^{-3}$ for gross-$\beta$ activity. A range of $6.1 \times 10^{-3}$ Bq m$^{-3}$ and the mean value of $4.4 \times 10^{-3}$ Bq m$^{-3}$ at ground level air were measured for $^7$Be activity.

This latter value is consistent with concentrations of $^7$Be in ground level air that have been reported by others. For 28 sampling sites, Feely et al. (1989) listed the mean $^7$Be surface air concentration for each month, averaged over all sampling years. The yearly arithmetic mean registered at our sampling site for 1993 and 1994 were $(4.3 \pm 1.4) \times 10^{-3}$ and $(4.4 \pm 1.4) \times 10^{-3}$ Bq m$^{-3}$, respectively, indicate that production rates of this radionuclide is relatively constant with time and undergoes approximately the same annual variations which are mainly due to the same cause: the variations in the intensity of vertical.

<table>
<thead>
<tr>
<th>Data</th>
<th>Gross $\alpha$ (Bq m$^{-3}$)</th>
<th>Gross $\beta$ (Bq m$^{-3}$)</th>
<th>$^7$Be (Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AM</td>
<td>0.75</td>
<td>5.68</td>
<td>4.4</td>
</tr>
<tr>
<td>S.D.</td>
<td>0.33</td>
<td>2.39</td>
<td>1.4</td>
</tr>
<tr>
<td>GM</td>
<td>0.68</td>
<td>5.20</td>
<td>4.1</td>
</tr>
<tr>
<td>DF</td>
<td>1.54</td>
<td>1.53</td>
<td>1.4</td>
</tr>
<tr>
<td>Maximum</td>
<td>2.03</td>
<td>13.09</td>
<td>7.7</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.16</td>
<td>0.92</td>
<td>1.5</td>
</tr>
</tbody>
</table>
turbulent exchange in the troposphere (Gedeonov et al., 1969; Feely et al., 1989).

3.1. Temporal variations of gross \(\alpha\), gross \(\beta\) and \(^{7}\text{Be}\) concentrations

Table 2 lists the mean concentrations (GM concentration for gross \(\alpha\) and gross \(\beta\) activities and AM concentration for \(^{7}\text{Be}\)) and their corresponding error (dispersion factor and standard deviation, respectively), for each month, averaged over all sampling years for all samples collected at our site. The number of samples in each mean \((n)\) is also given.

On examining this table, it can be said:

(a) With regard to the monthly GM concentrations, the higher values are those of July and August for both activities.

(b) The lowest values of gross-\(\alpha\) activity are registered in April and October while those of gross-\(\beta\) activity came from April and January.

(c) The data of our sampling site show a seasonal variation in concentrations of \(^{7}\text{Be}\). Seasonal variations show minimum averages in fall and winter months and maximum averages during the summer months probably reflecting the seasonal variations in the transport rate of air from the stratosphere to the troposphere.

The \(^{7}\text{Be}\) data for most of the other sites at a similar latitude in northern hemisphere show a pronounced seasonal variation in surface air concentrations of this radionuclide. Such seasonal variations have long been known to occur (Cruikshank et al., 1956; Parker, 1962; Peirson, 1963; Schumann and Stoeppler, 1963; Marenco and Fontan, 1967). Observed seasonal variations in the concentration of \(^{7}\text{Be}\) in surface air have often been attributed to the influence of variations in the exchange rate of air between the stratosphere and the troposphere. Although this influence is clearly real, other factors are also of importance. At sites in middle latitudes, variations in concentration of \(^{7}\text{Be}\) also result in part from some seasonal variations in the rate of vertical mixing within the troposphere with the highest concentrations being found during the warmer months (Viezee and Singh, 1980; Dutkiewicz and Husain, 1985).

We have plotted the weekly concentrations to compare the behaviors of the nuclides over the period of collection. The data we present for illustration in Fig. 5 are those from 1994. According to Gold et al. (1964) changes in \(^{210}\text{Pb}\) and \(^{7}\text{Be}\) levels often varied in the same pattern as gross fission product activity and specifically those for the long-lived fission product \(^{137}\text{Cs}\) despite their different origins and their different distribution throughout the atmosphere. Feely et al. (1989) attempted to evaluate the impact to the rate of transport of stratospheric air into the troposphere on the variations in surface air of \(^{7}\text{Be}\) by comparing these variations to those in surface air concentrations of a fission product, such as \(^{137}\text{Cs}\). When stratospheric air is carried into the troposphere – and this is most common at mid latitudes and most rapid during the spring season – fission product such as \(^{137}\text{Cs}\) as well as cosmogenic nuclides such as \(^{7}\text{Be}\)
Table 2  
Mean monthly measured concentrations in surface air averaged over all sampling years

<table>
<thead>
<tr>
<th>Month</th>
<th>Gross α ($ \times 10^{-4} $ Bq m$^{-3}$)</th>
<th>Gross β ($ \times 10^{-4} $ Bq m$^{-3}$)</th>
<th>$^7$Be ($ \times 10^{-3} $ Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GM</td>
<td>DF</td>
<td>n</td>
</tr>
<tr>
<td>Jan.</td>
<td>0.66</td>
<td>1.7</td>
<td>18</td>
</tr>
<tr>
<td>Feb.</td>
<td>0.67</td>
<td>1.5</td>
<td>16</td>
</tr>
<tr>
<td>Mar.</td>
<td>0.73</td>
<td>1.6</td>
<td>16</td>
</tr>
<tr>
<td>Apr.</td>
<td>0.57</td>
<td>1.9</td>
<td>17</td>
</tr>
<tr>
<td>May.</td>
<td>0.61</td>
<td>1.4</td>
<td>18</td>
</tr>
<tr>
<td>Jun.</td>
<td>0.61</td>
<td>1.3</td>
<td>16</td>
</tr>
<tr>
<td>Jul.</td>
<td>0.94</td>
<td>1.3</td>
<td>13</td>
</tr>
<tr>
<td>Aug.</td>
<td>0.95</td>
<td>1.4</td>
<td>13</td>
</tr>
<tr>
<td>Sep.</td>
<td>0.76</td>
<td>1.5</td>
<td>12</td>
</tr>
<tr>
<td>Oct.</td>
<td>0.57</td>
<td>1.5</td>
<td>13</td>
</tr>
<tr>
<td>Nov.</td>
<td>0.67</td>
<td>1.4</td>
<td>13</td>
</tr>
<tr>
<td>Dec.</td>
<td>0.65</td>
<td>1.4</td>
<td>12</td>
</tr>
</tbody>
</table>

Fig. 5. Weekly variations of gross α, gross β and $^7$Be in ground level air during 1994.

are transported into the upper layers of the troposphere. Once in the troposphere, the $^{137}$Cs will be affected by the same processes that affect the $^7$Be. For our sampling site, the peaks in the weekly concentration values for gross β and $^7$Be show similar changes, specially during the warmer months. This relation appears to be affected by meteorological conditions so once this has been achieved, the factors affecting concentrations in air have been studied.

3.2. Analyses of some meteorological factors affecting variation in concentrations

The study of meteorological fluctuations has reached, in the last few years, an increasing importance to environmental pollution researchers because its knowledge permits the elaboration of empirical models able to predict periods of potential radioactive pollution. As meteorology plays an important role in the dispersion and
transport of pollutants, we have performed a study to identify which meteorological parameters are strongly associated with the fluctuations of weekly concentrations. During the period of this study, meteorological data (wind direction, wind speed, temperature, pressure, relative humidity, precipitation and sunshine hours) were supplied by the nearby weather station (Málaga Airport Observatory).

First, we analyzed the influence of wind direction on the gross $\alpha$, gross $\beta$ and $^7\text{Be}$ concentrations. Málaga is on the Mediterranean coast and it is surrounded by mountains to the north. Due to the influence of the local orography there exists a prevailing SE wind and NW wind (Ortega y Sánchez, 1976). The SE and NW occur for sea–land and land–sea breezes, respectively. Strong winds in conjunction with easterly regime at Málaga are generally associated with disturbances and precipitation. This regime of winds has a direct effect on atmospheric aerosol concentrations. Studies of the influence of the nature of air-masses on the concentrations of radionuclides such as $^{222}\text{Rn}$ and its daughters and $^{212}\text{Pb}$ are effectively higher for NW winds than SE winds (Dueñas et al., 1990, 1994).

To study the potential influence of wind direction we have only taken into account weekly wind directions higher than 50% accordingly to the weekly activity values in order to avoid overlapping of the different regimes. The data of wind direction were also grouped by a wind velocity less than 3 m s$^{-1}$ so as to make the wind dispersion effect negligible and the average activity of each group was calculated (Table 3).

In the present study the gross $\alpha$ activity when westerly winds dominate are about as high as those from SE although they drop to rather low values when the winds have an easterly component. We believe this is due to the measurement being carried out weekly and the frequency of wind direction has been averaged accordingly to the sampling period, masking the effect of the continental or oceanic origin. For gross $\beta$ activity also, the differences in average concentrations can be considered to be negligible. $^7\text{Be}$ concentrations in ground level air are independent of the prevailing wind directions at Málaga, confirming the cosmogenic origin of $^7\text{Be}$ and its concentrations not being derived from a source of either oceanic or continental origin.

Next we studied the variability of the gross $\alpha$, gross $\beta$ and $^7\text{Be}$ concentrations at this station as resulted from local meteorological conditions. First, we performed a simple regression of the gross $\alpha$, gross $\beta$ and $^7\text{Be}$ concentrations and some meteorological factors and then we carried out a multiple regression in order to determine the extent to which the variations in concentrations might be attributed to the combination of these meteorological parameters. In our analysis we use weekly average of the daily maximum temperature ($T_{\text{maxmed}}$), weekly precipitation (LL), weekly average of the relative humidity ($H_{\text{med}}$), weekly hours of sunshine ($I$), weekly average pressure ($P_{\text{med}}$) and weekly average wind speed ($V_{\text{med}}$).

In Table 4, the correlation coefficient between concentrations and those meteorological factors are summarized. None of these correlation coefficients are definitive but provide limited fundamental insight. A weak correlation coefficient does not necessarily imply the absence of a causative relation, just that if a relation exists and it is linear, it is a relatively small effect in terms of explaining the major variations in the data.

The study of the correlations for gross $\alpha$ revealed a pronounced negative correlations with $V_{\text{med}}$ and LL. The next most important was a positive one with $P_{\text{med}}$. Weaker, but significantly, were seen with $T_{\text{maxmed}}$ and $H_{\text{med}}$ and lesser correlation with $I$. Meteorological

### Table 3
Mean values of gross $\alpha$, gross $\beta$ and $^7\text{Be}$ according to wind direction

<table>
<thead>
<tr>
<th>Wind direction</th>
<th>Gross $\alpha$ ($\times 10^{-4}$ Bq m$^{-3}$)</th>
<th>Gross $\beta$ ($\times 10^{-4}$ Bq m$^{-3}$)</th>
<th>$^7\text{Be}$ ($\times 10^{-3}$ Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NW</td>
<td>$0.9 \pm 0.3$</td>
<td>$6.3 \pm 2.1$</td>
<td>$4.3 \pm 1.4$</td>
</tr>
<tr>
<td>SE</td>
<td>$0.8 \pm 0.3$</td>
<td>$6.4 \pm 2.9$</td>
<td>$4.2 \pm 1.4$</td>
</tr>
</tbody>
</table>

### Table 4
Linear correlation coefficient between surface air concentrations and some meteorological factors

<table>
<thead>
<tr>
<th>Activity</th>
<th>$T_{\text{maxmed}}$</th>
<th>LL</th>
<th>$H_{\text{med}}$</th>
<th>$I$</th>
<th>$P_{\text{med}}$</th>
<th>$V_{\text{med}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gross $\alpha$</td>
<td>0.18</td>
<td>$-0.27$</td>
<td>0.18</td>
<td>0.10</td>
<td>0.24</td>
<td>$-0.31$</td>
</tr>
<tr>
<td>Gross $\beta$</td>
<td>0.34</td>
<td>$-0.27$</td>
<td>0.17</td>
<td>0.12</td>
<td>0.15</td>
<td>$-0.39$</td>
</tr>
<tr>
<td>$^7\text{Be}$</td>
<td>0.38</td>
<td>$-0.25$</td>
<td>$-0.17$</td>
<td>0.38</td>
<td>0.03</td>
<td>$-0.12$</td>
</tr>
</tbody>
</table>
variables that correlated with gross $\beta$ activity were in order of importance: $V_{med}$, $T_{maxmed}$, LL, $H_{med}$, $P_{med}$ and $I$. Judging from the linear coefficient values, it can be said that a high concentration of $^{7}$Be is present under conditions of high temperature ($T_{maxmed}$) and hours of sunshine ($I$). There is strong negative correlation between the data and precipitation and a lesser correlation with $H_{med}$, $V_{med}$ (negative) and $P_{med}$ (positive) do not seem to have any distinct effect.

In order to find the meteorological factors that influence the gross $\alpha$, gross $\beta$ and $^{7}$Be concentrations and evaluate them in order of importance, a multiple regression analysis was carried out with the STATGRAPHICS-PLUS program. The meteorological variables were obtained chosen by decreasing order of the linear coefficient. Tables 5–7 present by columns: the regression equations with the error of the coefficient of each independent variable, the $R$-squared value (the amount of the variance accounted for by the model), the probability index ($p$) taking into account the number of data and the standard error of the estimate (S.E.) (the square root of the residual mean square).

In the validity analysis of each one of the regression equations were taken into account: the relative error of the coefficient of each independent variable, the standard error of the estimate and the $R$-squared value. Using these criteria, the following equations were chosen for the gross $\alpha$ and gross $\beta$, respectively, from the results of the

### Table 5
Model fitting results for gross $\alpha$ activity

<table>
<thead>
<tr>
<th>Regression equations</th>
<th>$R$</th>
<th>$p$ (%)</th>
<th>S.E.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_\alpha(\cdot 10^{-4}) = (1.07 \pm 0.08) - (0.092 \pm 0.022)V$</td>
<td>0.31</td>
<td>&gt; 99</td>
<td>0.31</td>
</tr>
<tr>
<td>$A_\alpha(\cdot 10^{-4}) = (1.16 \pm 0.08) - (0.103 \pm 0.021)V - (0.0076 \pm 0.0017)LL$</td>
<td>0.44</td>
<td>&gt; 99</td>
<td>0.30</td>
</tr>
<tr>
<td>$A_\alpha(\cdot 10^{-4}) = -(12 \pm 5) - (0.105 \pm 0.020)V - (0.0065 \pm 0.0017)LL + (0.013 \pm 0.005)P$</td>
<td>0.48</td>
<td>&gt; 99</td>
<td>0.29</td>
</tr>
<tr>
<td>$A_\alpha(\cdot 10^{-4}) = -(24 \pm 6) - (0.092 \pm 0.020)V - (0.0033 \pm 0.0019)LL + (0.024 \pm 0.006)P + (0.018 \pm 0.006)T$</td>
<td>0.52</td>
<td>&gt; 99</td>
<td>0.28</td>
</tr>
<tr>
<td>$A_\alpha(\cdot 10^{-4}) = -(23 \pm 6) - (0.07 \pm 0.03)V - (0.004 \pm 0.002)LL + (0.023 \pm 0.006)P + (0.021 \pm 0.006)T + (0.004 \pm 0.004)H$</td>
<td>0.52</td>
<td>&gt; 99</td>
<td>0.28</td>
</tr>
</tbody>
</table>

### Table 6
Model fitting results for gross $\beta$ activity

<table>
<thead>
<tr>
<th>Regression equations</th>
<th>$R$</th>
<th>$p$ (%)</th>
<th>S.E.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_\beta(\cdot 10^{-4}) = (8.7 \pm 0.6) - (0.87 \pm 0.15)V$</td>
<td>0.39</td>
<td>&gt; 99</td>
<td>2.21</td>
</tr>
<tr>
<td>$A_\beta(\cdot 10^{-4}) = (5.2 \pm 1.0) - (0.78 \pm 0.15)V + (0.14 \pm 0.03)T$</td>
<td>0.49</td>
<td>&gt; 99</td>
<td>2.10</td>
</tr>
<tr>
<td>$A_\beta(\cdot 10^{-4}) = (6.7 \pm 1.0) - (0.87 \pm 0.14)V + (0.10 \pm 0.03)T - (0.046 \pm 0.012)LL$</td>
<td>0.54</td>
<td>&gt; 99</td>
<td>2.03</td>
</tr>
<tr>
<td>$A_\beta(\cdot 10^{-4}) = -(0.56 \pm 0.22)V + (0.14 \pm 0.04)T - (0.049 \pm 0.012)LL + (0.05 \pm 0.03)H$</td>
<td>0.56</td>
<td>&gt; 99</td>
<td>2.01</td>
</tr>
</tbody>
</table>

### Table 7
Model fitting results for $^{7}$Be

<table>
<thead>
<tr>
<th>Regression equations</th>
<th>$R$</th>
<th>$p$ (%)</th>
<th>S.E.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{Be-\alpha}(\cdot 10^{-3}) = (2.0 \pm 0.5) + (0.106 \pm 0.023)T$</td>
<td>0.38</td>
<td>&gt; 99</td>
<td>1.27</td>
</tr>
<tr>
<td>$C_{Be-\alpha}(\cdot 10^{-3}) = (1.8 \pm 0.5) + (0.07 \pm 0.03)T + (0.019 \pm 0.100)I$</td>
<td>0.42</td>
<td>&gt; 99</td>
<td>1.25</td>
</tr>
<tr>
<td>$C_{Be-\alpha}(\cdot 10^{-3}) = (2.1 \pm 0.6) + (0.07 \pm 0.03)T + (0.016 \pm 0.010)I - (0.008 \pm 0.010)LL$</td>
<td>0.42</td>
<td>&gt; 99</td>
<td>1.25</td>
</tr>
<tr>
<td>$C_{Be-\alpha}(\cdot 10^{-3}) = (0.9 \pm 1.4) + (0.06 \pm 0.03)T + (0.021 \pm 0.012)I - (0.009 \pm 0.010)LL + (0.013 \pm 0.015)H$</td>
<td>0.43</td>
<td>&gt; 99</td>
<td>1.25</td>
</tr>
</tbody>
</table>
stepwise backward regression method:

\[
A_a(10^{-4}) = -(12 \pm 5) - (0.105 \pm 0.020)V
- (0.0065 \pm 0.0017)LL + (0.013 \pm 0.005)P, \quad (1)
\]

\[
A_b(10^{-4}) = -(6.7 \pm 1.0) - (0.87 \pm 0.14)V
+ (0.10 \pm 0.03)T + (0.046 \pm 0.012)LL. \quad (2)
\]

These equations exhibit the number of parameters that most interfere in the fluctuations of the gross \( a \) and gross \( b \). Judging from these equations it can be said that the meteorological variables most influencing the weekly gross \( a \) activity are: wind speed, weekly precipitation and the weekly average pressure. For the weekly gross \( b \) activity are: the wind speed, the \( T_{\text{maxmed}} \) and the precipitation of that week.

With respect to \( ^{7}\text{Be} \), adding the \( I \) term to the model and the following terms in order, resulted in a relatively small rise in S.E. and \( R \) values. Besides the relative error of the coefficient of each independent variable increase considerably. This suggests that we could drop it from the model. According to the selected criteria, the final form of the model with meteorological variables for \( ^{7}\text{Be} \) concentrations is given in Eq. (3).

\[
C_{\text{Be}_a} = (10^{-3}) = (2.0 \pm 0.5) + (0.106 \pm 0.023)T. \quad (3)
\]

It can be said for \( ^{7}\text{Be} \) concentrations that our results showed a prevailing influence of \( T_{\text{maxmed}} \) above any other meteorological parameter for our period of sampling and site.

3.3. Model as a function of meteorological parameters

To test the validity of these equations in the estimation of gross \( a \) and gross \( b \) activities and \( ^{7}\text{Be} \) concentrations in Málaga, we performed a study applying Eqs. (1)–(3) to weekly data which were not used in the stepwise backward regression method. These data correspond to weeks from June 1995 to December 1995 (25 data). The study was conducted comparing the theoretical concentrations to that experimentally obtained.

In Figs. 6–8 we plotted the theoretical line (dotted line), which showed a concordance between the calculated values and those experimentally observed. The solid lines correspond to lineal least-squares fit to the data. The regression equations obtained are given as

\[
\alpha_{\text{observed}} (\text{Bq m}^{-3}) = (0.8 \pm 0.2)\alpha_{\text{calculated}}
+ (1.3 \pm 1.6) \times 10^{-5} \quad r = 0.64,
\]

\[
\beta_{\text{observed}} (\text{Bq m}^{-3}) = (1.3 \pm 0.3)\beta_{\text{calculated}}
+ (1.8 \pm 1.4) \times 10^{-4} \quad r = 0.71,
\]

\[
^{7}\text{Be}_{\text{observed}} (\text{Bq m}^{-3}) = (2.1 \pm 0.5)^{7}\text{Be}_{\text{calculated}}
- (4.7 \pm 2.4) \times 10^{-3} \quad r = 0.69,
\]

The correlation coefficients \( r \) 0.64, 0.71 and 0.69 were significant at a probability index \( p > 99\% \). Our study revealed the validity of the relationship for gross \( a \) and gross \( b \) and the meteorological variables implied in the similarity in the graph slopes in Figs. 6 and 7,
respectively. It can be seen that there is a good agreement with data predicted from Eqs. (1) and (2) with those measured in the samples (Liger, 1996). From Fig. 8 it can be observed that the rough variation of \(^{7}\text{Be}\) concentrations may be explained on the basis of temperature differences. This is consistent with the fact that the solar heating of the surface of the Earth leads to the heating of the air in contact with the surface. Cooler air sinks, displacing the warm less dense air and forcing it upward. This new air is heated in turn and is forced upward. A convective circulation is produced, carrying surface air upward and bringing downward air from higher levels. This vertical transport carries down, to the surface layer, the \(^{7}\text{Be}\) that has been produced within the upper troposphere (Baskaran, 1995). Although, there are some systematic deviations between the \(^{7}\text{Be}\) concentrations in air and the corresponding values calculated from Eq. (3) especially when they are extreme: very high or very low. Great care must be taken in the interpretation of such differences. The highest concentrations during the last months of the year 1995 were registered particularly in summer months, especially in July, with a maximum value of \(7.9 \times 10^{-3} \text{ Bq m}^{-3}\). The results appear to indicate a considerable contribution of stratospheric origin to the \(^{7}\text{Be}\) at Málaga. At sites in mid latitudes, variations in the concentration of \(^{7}\text{Be}\) also result, in part, from seasonal variations in the rate of vertical mixing within the troposphere, with the highest concentrations being found during the warmer months. On the other hand, the lowest \(^{7}\text{Be}\) levels found at our sampling site correspond to predicted values somewhat higher. This leads us to think that in the prediction Eq. (3) another term function of a meteorological variable inversely related with \(^{7}\text{Be}\), such as rain, should appear. The parameter is itself of interest even if it has little impact on the predictive power of the model. This phenomenon is not easily explained. The generally accepted belief is that deposition of this
radionuclide occurs primarily in precipitation except in areas where precipitation is light (Young and Silker, 1980). Discussions of various sources suggested in recent technical literature have shown that several sites situated in climatic zones where there are strong seasonal variations in rainfall rate experience an inverse relationship between the rainfall rate and the $^7\text{Be}$ concentrations, especially indicating the importance of a washout of the atmospheric aerosol that carries $^7\text{Be}$ (Feely et al., 1989; Wallbrink and Murray, 1994; Ishikawa et al., 1995). The rainfall rates at those zones are four to seven times the annual average at Málaga. Another factor that should be considered is the variation of amounts in precipitation at Málaga that has shown a definite minimum in the last few years. The total annual rainfall during the 1992–1995 period range between 186.02 and 256.9 mm which are distributed throughout the year although summers tended to be even drier that the rest of the seasons. These amounts represents only 32 and 45% of the rainfall average at Málaga. However, this time period is not an exceptional situation for our sampling site. The 46% of the last 56 years were under the annual rainfall average at Málaga (Ortega and Sánchez, 1976).

A significant feature of the present results is that, although it seems to be masked by that of temperature in the regression process, this phenomenon can be qualitatively appreciated in Fig. 9 which represents weekly $^7\text{Be}$ concentration and weekly rainfall data during 1994. Rainfall being incidentally very strong or very light may result in rather extreme activity values. Although this effect is clearly noticeable in individual cases it does not play an important role in the overall evaluation of $^7\text{Be}$ owing to the relatively few rainfall events encountered at the site in which our experiments were conducted.

Although the rough variation of $^7\text{Be}$ concentrations is closer to the temperature pattern, the reason for the difference in the activity concentrations has yet to be clarified. Even though there is a stronger correlation with the temperature as compared to the amount of precipitation, it is quite likely this is one of the master variables that controls the amount of $^7\text{Be}$ in air. Although the model was not created for forecasting and cannot be still used for this purpose, the results are encouraging for building a forecasting model enabling us to predict periods of potential radioactive pollution in this zone.

References


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