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Studies on radioactivity in aerosols as a function of meteorological variables in Salamanca (Spain)

M. García-Talavera^{a,*}, B. Quintana^a, E. García-Díez^b, F. Fernández^a

^aGrupo de Física Nuclear, Universidad de Salamanca, E-37008 Salamanca, Spain

^bDpto. de Física de la Atmósfera, Universidad de Salamanca, E-37008 Salamanca, Spain

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Abstract

Ambient concentrations of aerosol radioactivity in ground-level air were measured weekly in a low-industrialized city (Salamanca, Spain). Means of about 6.9×10^{-5} and $4.83 \times 10^{-4} \text{ Bq m}^{-3}$ were found for gross α and β activities, respectively, averaged over three years. The measured activities, which include contributions from several radionuclides in the ground atmosphere, were evaluated to determine the relationship between the meteorology and the aerosol activities in air. We have studied the influence of diverse meteorological parameters such as temperature, pressure and wind direction, as well as the effect of the less often considered terrestrial electrostatic field. Concentrations of gross α and β activities were greatly affected by the meteorological conditions, showing pronounced differences between seasons. The study reveals that much of the variability (approximately 50%) in the activities is explained by the electrostatic field, its influence being higher in winter. About 40% of the α and β variability is explained by both the temperature and the humidity. The influence of the wind direction is only statistically significant for α radioactivity. A simple mathematical model based on these variables is developed to describe globally the dynamics in air of radioactive particles enabling us to estimate the air radioactive background level. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The terrestrial atmosphere is the main path through which the radioactivity is transmitted in the environment. Therefore, the knowledge of the mechanisms that make the airborne radioactivity vary is needed to estimate its effect on a specific ecosystem. But to fully understand the processes responsible for its spatial and temporal distribution requires analysis of the effect that meteorology has on the activity levels. Regional meteorology influences mainly the spatial distribution, while for studying the temporal distribution in a specific place, local meteorology has to be considered. This paper examines the temporal relationship between meteorological

parameters and activity concentrations. Therefore, only local atmospheric variables are involved in this work.

Concentration of gases and particulate matter have been proved to be affected by the meteorological variables over a wide range of environments, from urban locations (Sequeira and Lai, 1998) to high mountain sites (Kasper and Puxbaum, 1998). Several studies have also been performed on the relationship between the meteorological parameters and the concentration of diverse radionuclides in air. Various factors such as temperature (Kolb, 1989), atmospheric stability (Ranjarajan et al., 1986) or precipitation (Fujitaka et al., 1992; Nishikawa et al., 1988) seem to be related to the activity values.

A less-studied influence is the one that the atmospheric electrostatic field exerts on the activity levels. The electric field existing between the terrestrial surface and the stratosphere causes an increase of the positive ions near

* Corresponding author.

the ground which acts as the negative electrode (Israel and Dolezalek, 1971). The fact that radon decay products, which constitute the major source of radioactivity in air (De Cort et al., 1997), begin their existence as positive ions and remain positively charged or combine with other atoms to provide clustering with polar molecules, makes the atmospheric field play an important role in airborne radioactivity. At the same time, radon and its descendants affect the electric field, being an important component of the electric properties of the atmosphere (Wilkening, 1990).

Radioactivity levels in air should be controlled, because α and β particles are hazardous to humans when emitter nuclei are breathed. The characterization of these levels can be done in terms of the number of α and β emissions produced per unit of time and air volume (gross α and β activities), which are determined from the sampling of atmospheric aerosols. Systematic measurement of aerosol gross activities enables us to study periodic variations and the main meteorological causes influencing the environmental radioactivity levels, besides allowing to detect activity peaks.

In this paper, we investigate those factors affecting the temporal distribution of the radioactive aerosol concentrations by measuring weekly the gross α and β activities, during three years (1993–1995) at Salamanca city (Spain). In this town, the local anthropogenic sources are practically limited to coal combustion for heating and emissions from the automobiles in use. These factors together with the natural sources constitute the local environmental radioactive background. We study its dependence with respect to different atmospheric variables and the electric field, and propose two regression models which describe the α and β activity behavior as a function of these variables. It allows to estimate quantitatively the importance of each of the meteorological variables in the variability of the activity in air. Besides, the development of such models is the first step to detect significant increments of the usual values. Both models have been validated using activity data that were not included to calculate the fitting parameters.

2. Sampling and methods of analysis

The aerosol samples were collected at a height of 15 m above ground in Salamanca (40°56'N, 5°29'W), a medium size city (169,000 inhabitants) mainly dedicated to agricultural and cattle raising activities. The sampling that we have established is continuous and systematic. It consists of collecting the samples according to a given temporal distribution. In our case, the time interval is weekly. However, the sampling is continuous in order to detect possible peaks of radioactivity (Gilbert, 1987). The sampler is a pump, model Raddeco AV-28A. It makes air flow through a cellulose membrane filter of pore size

0.8 μm with a constant velocity of 30 cm s^{-1} . The filter efficiency is very high for all sizes of particles, reaching a minimum value of 40% for 0.1 μm diameter particles (Hinds, 1982).

Dust contents were determined by weighting the filters before exposure and four days after exposure under the same conditions of temperature and humidity. Thus, the shortest half-life elements will have decayed at the start of the measurement. The gross α activity is measured with a ZnS(Ag) scintillation counter whose associated electronic chain is composed by a Canberra Model 2007P photomultiplier tube, a Canberra Model 2015A AMP/TSCA amplifier and a Canberra Model 2071 Dual Counter Timer. The gross β activity is recorded by means of a proportional counter (Canberra Model 2404). Because of the low activity of the filter samples we settle an acquiring time of 600 min, long enough to obtain relative uncertainties around 10%. The resulting detection limits are $1.68 \times 10^{-7} \text{ Bq m}^{-3}$ for α activity measurements and $2.42 \times 10^{-6} \text{ Bq m}^{-3}$ for β activity measurements.

Measurements by γ spectrometry were performed to determine the ^{210}Pb activity of the samples. Due to the higher detection limits of this technique, the measurements could not be performed on single filters, but on samples composed of 12 filters each. The γ spectrometer is an n-type HPGe detector, whose active volume is 117 cm^3 and specifications are relative photopeak efficiency at 1332 keV of 28.3% and resolutions at 122 and 1332 keV of 0.860 and 1.87 keV, respectively. The associated preamplifier is a Canberra Model 2008. This is connected to an ORTEC Model 572 amplifier and this to a Canberra 8701 analog-digital converter. Spectra were stored through an AccuSpec/A interface card installed in a PC computer.

The meteorological data are provided by the Instituto Nacional de Meteorología, from an automatic station located in the southwest of the city. The electrostatic field is measured by an electric field mill manufactured by Atmospheric Research Systems Inc., which is placed next to the air sampler. Weather data are taken every 10 min and afterwards they are averaged weekly to fit them to the sampling periods of the filters.

3. Results and discussion

3.1. Sources and time behavior of the airborne gross α and β activities

Since 1993 until 1996, a sample per week has been measured for gross α activity (A_α) and gross β activity (A_β) determinations. The measurement uncertainty has been estimated to be 10.7% for α counting and 7.3% for the β one. In Fig. 1, the time behavior of the α and β measurements along the three studied years is represented together with the aerosol content per sampled air

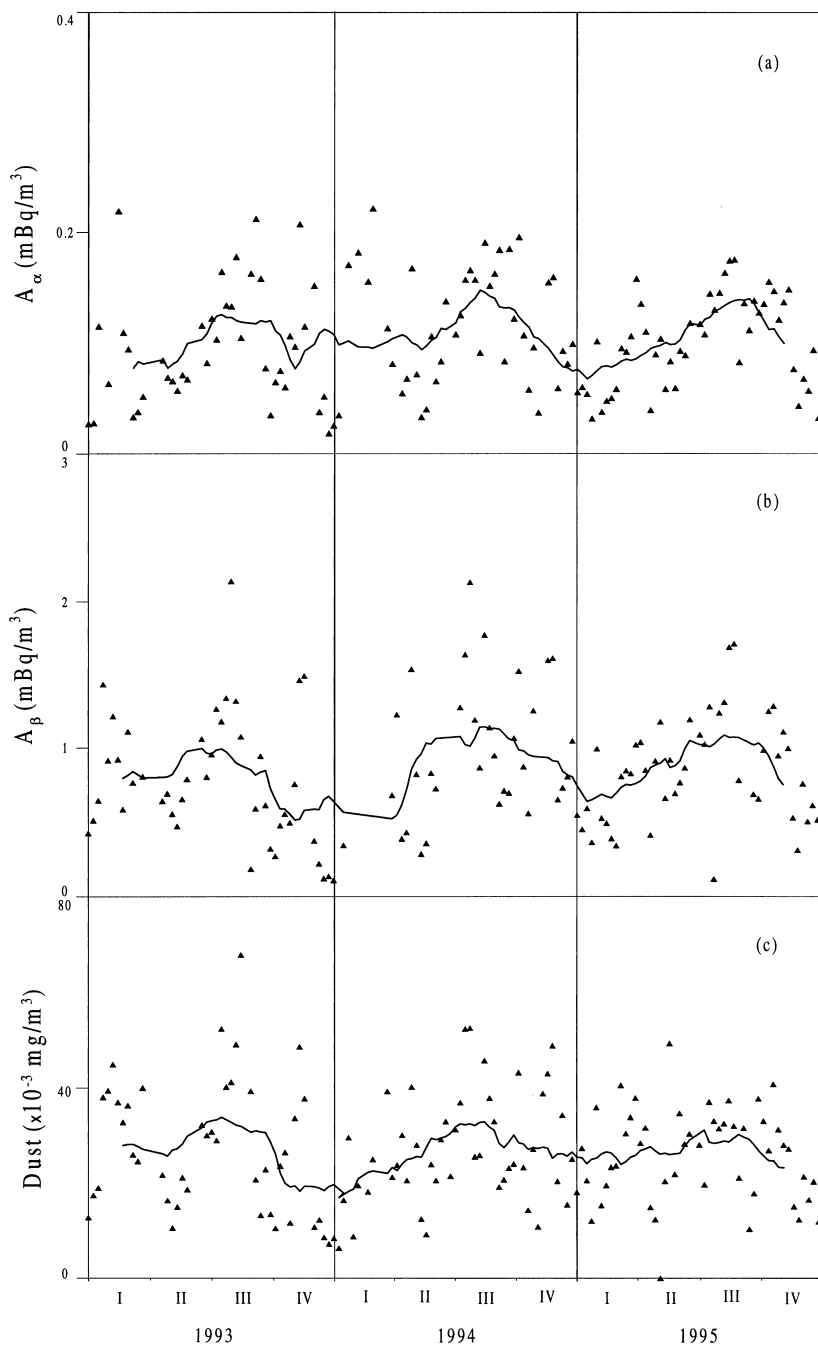


Fig. 1. (a) Gross α activity distribution along the three studied years. The solid line represents a smoothing of data achieved for a better observation of the seasonal trend. (b) As in (a) but for gross β activity. (c) As in (a) but for the dust content of the air.

volume of the filters. The mean values determined over these three years are 6.9×10^{-5} and $4.83 \times 10^{-4} \text{ Bq m}^{-3}$, respectively, and the minimum detectable activity above these environmental levels are 4.0×10^{-6} and $2.5 \times 10^{-5} \text{ Bq m}^{-3}$. The specific α activity of the samples is on average 4.0 Bq g^{-1} and the β activity, 31.7 Bq g^{-1} .

As can be seen in Table 1, the major source of gross β activity is ^{210}Pb , which is a long-lived descendant of gaseous ^{222}Rn . This result can be justified in the light of two facts: atmospheric radioactivity is known to be dominated by the naturally occurring short-lived particulate decay products of gaseous radon, and; in view of the

Table 1

Gross β and ^{210}Pb activities of composite filter samples together with the percentage of the gross β activity accounted for ^{210}Pb . The measurement uncertainties are given in brackets. The low percentage corresponding to the sampling period 01.07.94–30.09.94 make us suspect that the ^{210}Pb activity was wrongly determined

Sampling period	A_{β} ($\times 10^{-4}$ Bq m $^{-3}$)	A_{Pb} ($\times 10^{-4}$ Bq m $^{-3}$)	(%)
25.02.93–02.07.93	4.1 (3)	3.8 (1)	93 (7)
01.10.93–03.01.94	3.2 (3)	3.4 (1)	100 (10)
03.01.94–29.03.94	3.4 (2)	2.8 (2)	82 (8)
29.03.94–01.07.94	4.8 (3)	4.2 (2)	88 (7)
01.07.94–30.09.94	7.1 (4)	2.3 (2)	32 (3)
30.09.94–29.12.94	6.2 (4)	4.4 (2)	71 (6)
29.12.94–31.03.95	3.6 (2)	2.6 (2)	72 (7)
31.03.95–30.06.95	5.5 (3)	3.8 (1)	69 (4)
30.06.95–03.10.95	6.7 (4)	5.5 (2)	82 (6)
03.10.95–26.12.95	6.5 (4)	4.9 (1)	75 (5)

4-days decay period allowed for the filters before counting, the shortest-lived radon descendants would have decayed to negligible levels. This means that only ^{210}Po would contribute significantly to the measured gross α activity, while ^{210}Pb and to a less extent ^{210}Bi , with half-life 5.1 days, would contribute to the gross β activity. Nonetheless, ^{210}Po and ^{210}Bi concentrations in the atmosphere are much lower than that of its ancestor, ^{210}Pb . The reason for this disequilibrium is that ^{210}Pb half-life (22.1 yr) is very long compared to its atmospheric residence time, which is about 8 days. Values of the ratio $^{210}\text{Po}/^{210}\text{Pb}$ measured at various locations around the world (Chamberlain, 1991) resembles the average ratio A_{α}/A_{β} of the filters collected at Salamanca. This lead us to assess that ^{210}Po constitute the main source of α activity. However, other secondary sources of radioactivity, such as resuspended soil particles or anthropogenic activities, must be taken into account. The contribution of the former will be limited to strong wind periods, because otherwise it is not probable that coarse soil particles ($< 2 \mu\text{m}$) reach the air sampler, placed at 15 m height.

Regarding anthropogenic radioactivity, the most relevant source in the place of study is coal burning, which is limited to the cold period. According to the values reported by Beck and Miller, 1980, we have estimated the specific α and β activities of coal fly ashes in 3.08 and 3.47 Bq g $^{-1}$, respectively. While the specific α activity is comparable to that of the aerosol samples, the specific β activity is an order of magnitude lower. Therefore, coal burning can only represent a significant contribution to the α radioactive level. This difference between the α and the β activities allows us to extract further conclusions from the analysis of the ratio A_{α}/A_{β} . The fact that this ratio has not increased during the cold period but displays a constant trend along the year, shows that on average the α activity level also is not significantly increased by coal fly ashes. However, an increment in coal

ashes could explain the episodic events detected at the end of the winter in 1993 and 1995, when unusually high values of the ratio A_{α}/A_{β} were detected for some filters.

According to the results discussed above, the contribution of anthropogenic sources, which could mask natural trends of the radioactive background, is not high. Out of this, a marked seasonal behavior is expected, because the mechanism of particle formation, dispersion and deposition, ruling the aerosol concentration in air, are strongly connected to the atmospheric conditions. But before searching for seasonal variations in the data, the activity distributions recorded each year were compared. As the annual data distributions are nonnormal, we have employed a nonparametric test which compares the medians of the statistical distributions for each year. Neither the gross A_{α} nor the A_{β} showed significant differences among years. From a visual inspection to the data (see Fig. 1), seasonal changes seem to be produced in a regular way for the three years. To compare the medians of each season (Table 2), we have employed the test of Mann–Whitney (Bunzl, 1993). The results given by the test for a 95% confidence level indicate that there are significant differences in the activity levels not only between summer and winter, but also between spring and summer, and between autumn and winter (Tables 3 and 4). However, the test values resulting from the comparison between summer and autumn, and winter and spring correspond to probabilities higher than 5% of being nondiffering distributions. Slight differences are obtained in the test for α and β radiation probably due to the different sensitivities of α and β measurements, but essentially the behavior is similar.

Considering the results given in Tables 2–4, we can conclude that the activity maxima are registered in summer and minima in winter. The activity also tends to be higher in autumn than in spring, although differences are only statistically significant for the α activity (Tables 3

Table 2

Medians (Bq m^{-3}) of the α and β activity statistical distributions corresponding to the four seasons

	Winter	Spring	Summer	Autumn
M_α	3.60×10^{-5}	4.42×10^{-5}	8.34×10^{-5}	6.51×10^{-5}
M_β	3.07×10^{-4}	4.59×10^{-4}	6.86×10^{-4}	4.79×10^{-4}

Table 3

Result of the Mann–Whitney test for the comparison between α -gross activity distributions of two seasons

	Spring	Summer	Autumn
Winter	– 1.048	– 4.330	– 2.679
Spring		– 5.651	– 2.682
Summer			– 2.455

Table 4

Same as in Table 3 but for the β -gross activity distributions

	Spring	Summer	Autumn
Winter	– 2.847	– 5.135	– 3.671
Spring		– 4.264	– 1.209
Summer			– 1.892

and 4). As these two seasons were quite similar in temperature and precipitation in the years of the study we can ascribe this fact to the behavior of cold and warm air masses. Cold masses are characterized by being at a lower temperature than the ground over the layout. When cold air masses start heating, the instability increases, developing vertical flows which disperse the suspended particles. In spring, the air behaves as a cold mass, because of the fast heating of the ground produced by sun exposure. However, in autumn it behaves as a warm mass due to the high radiative cooling of the ground, compelling the particles to accumulate in the lower layers (Jansa Guardiola, 1985).

The seasonal behavior of aerosol activities indicates a clear dependence on the climatological conditions. To further understand the last factors influencing this seasonal behavior, weather variables should be included in our studies.

3.2. Influence of the meteorological variables

To select the main variables affecting the activities, we carried out correlation studies. The monotonic increase or decrease of the activities with respect to the meteorological variables: temperature, humidity, pressure and precipitation, has been evaluated by the calculation of

Table 5

Correlation studies between α activity and the main meteorological variables. Values of Spearman (ρ) and Pearson (r) coefficients are given together with the associated correlation probabilities (p)

Variable	ρ (p)	r (p)
Temperature	0.359 (< 0.05)	0.507 (< 0.05)
Humidity	– 0.329 (< 0.05)	– 0.441 (< 0.05)
Pressure	– 0.053 (> 0.05)	– 0.106 (> 0.05)
Precipitation	– 0.330 (< 0.05)	– 0.191 (< 0.05)

Table 6

Correlation studies between β activity and the main meteorological variables

Variable	ρ (p)	r (p)
Temperature	0.450 (< 0.05)	0.548 (< 0.05)
Humidity	– 0.475 (< 0.05)	– 0.545 (< 0.05)
Pressure	– 0.143 (> 0.05)	– 0.175 (> 0.05)
Precipitation	– 0.421 (< 0.05)	0.184 (> 0.05)

the Spearman coefficient (Bunzl, 1993). As we can see in Tables 5 and 6, the pressure is the only variable not significantly correlated to the α and β activities at a 95% confidence level. Temperature and humidity are the variables most strongly correlated to the activities. High temperatures are often associated to upward convection currents in the atmosphere favoring radon emanation and dispersion of aerosol particles. This is reflected in a positive correlation between temperature and radioactive levels. On the contrary, the correlation with the humidity is negative. The reason is that high levels of humidity inhibit radon emanation by means of a decrease of soil porosity. The connection of the airborne radioactivity with these two meteorological variables explains why the highest activity values appear during the summer. We have also calculated the linear correlation by means of the Pearson coefficient (Tables 5 and 6). Temperature and humidity are the variables with a higher linearity. Precipitation and pressure are nonlinearly correlated.

The direction of the wind may be related to the gross activities too, since the origin of the transported particles

Table 7

Spearman coefficients for the activities and the electrostatic atmospheric field

Variable	ρ (p)
α activity	0.372 (< 0.05)
β activity	0.480 (< 0.05)

could make the fraction of radioactive aerosols vary. The northwest (NW) wind was prevailing in over a half of the analyzed weeks, and therefore the frequency of the NW wind was selected as a variable for the study. Besides, it is important to point out that the wind coming from this direction has crossed the city before reaching the sampler. The Spearman coefficients (ρ) indicate that while a significant positive correlation exists with the α activity (ρ of 0.34 for a 95% confidence level), there is not a considerable influence of the wind on the β activity. This is probably due to the higher detection level for the gross β activity. The high β level induced by ^{210}Pb coming from atmospheric radon makes the detection of radioactivity variations from other sources difficult. For this reason the gross β measurement is less sensitive than the gross α to changes in the origin of the transported particles.

3.3. Influence of the atmospheric electrostatic field

Once the influence of the meteorological variables has been evaluated, we have studied the effect of the atmospheric electric field on the radioactive aerosols. Since about the 88% of the radioactive emitters are positively charged as a result of the nuclear decay process (Marlow, 1988), the electric field could affect their pathway in the atmosphere. The measurements of mobilities of the positive radon decay product ions clearly show that the electrical mobilities, and hence the masses of the radon daughter positive ions fall in the same general range as those for ordinary atmospheric small ions, which are highly governed by the electric field. Indeed, various studies confirm that the charged ^{222}Rn products in the atmosphere are affected by the electric fields of thunderstorms, as well as by normal fair-weather electric fields (Wilkening, 1990). Furthermore, polar molecules, to which radon progeny tends to attach, are also sensitive to be affected by the atmospheric electric field (Pohl, 1978). It must also be considered that the atmospheric electric field is affected by the meteorological conditions, specially by atmospheric pressure and precipitation. Therefore, a correlation with the field could be partly induced by the common effect of some weather variable on both field and aerosols concentration.

The Spearman coefficients for the gross activities were computed (Table 7), showing the existence of a positive

correlation, i.e., high field values correspond to high concentrations of radioactive emitters. Let us note that higher correlation values are obtained compared to any of the studied meteorological variables. If we carry out a seasonal study, we can observe that a much better correlation with the electric field is found in winter than in summer. The Spearman coefficients are 0.65 and 0.53 for α and β activities in winter, while no significant correlation is found in summer. This phenomenon could be explained by considering the fact that the convection currents in the atmosphere take place mainly in summer. In winter, when the atmosphere is much more stable, the electrostatic forces predominate over the convective ones, and therefore, the field has a greater influence on the particles.

3.4. Multiple regression model

Attending to the established correlations, we have developed a suitable model to characterize the environmental activity level of the air as a function of variables measured irrespective of the activity. Among the several possibilities studied, we have reached as better models for α and β activities, expressed as mBq m^{-3} , the following ones:

$$A_{\alpha} = a_1(E)^{-1}E + a_2\left(\frac{T}{H}\right)^{-1}\frac{T}{H} + a_3(v_{\text{NW}})^{-1}v_{\text{NW}}, \quad (1)$$

$$A_{\beta} = b_1(E)^{-1}E + b_2\left(\frac{T}{H}\right)^{-1}\frac{T}{H}, \quad (2)$$

where E is the terrestrial electric field, T the temperature; H the relative humidity and v_{NW} the northwest wind frequency. For the equations to be independent on the physical units of the meteorological variables, we have introduced the average values within the period 1993–1995 (E , T/H and v_{NW}). Known the averaged values of the three meteorological variables within one week the corresponding gross activities can be estimated by means of the regression equations.

To obtain the values of the coefficients, we have calculated a least-squares fit to the data and then we have identified aberrant data points by examining the residuals. Finally, the fit has been recalculated without these points. This robust application of the classical theory eliminates the disproportionate influence of exceptional data points on the fit (Hodgin et al., 1982). The values of the best-fit parameters are $a_1 = 0.0308(73)\text{mBq m}^{-3}$, $a_2 = 0.0412(60)\text{mBq m}^{-3}$, $a_3 = 0.0209(62)\text{mBq m}^{-3}$, for the α activity model; and $b_1 = 0.268(35)\text{mBq m}^{-3}$, $b_2 = 0.301(39)\text{mBq m}^{-3}$, for the β activity one.

Both models are very simple, their dependence on the variables being linear. They may be understood as a balance of strengths which are represented by the different terms of the equations. The first term represents the electrostatic force exerted on the radioactive aerosols, the

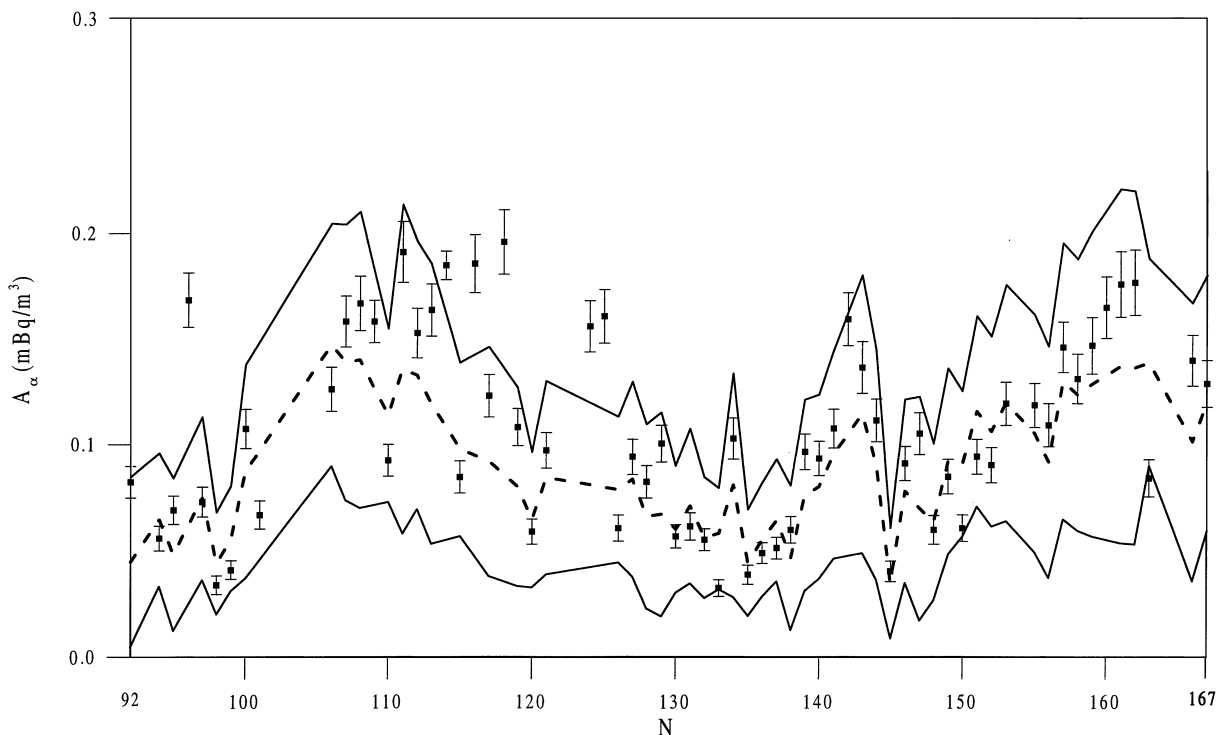


Fig. 2. Calculated activities of the α activity model. The dashed line represents the average and the solid lines the confidence interval at the 95%. The experimental data are represented by squares. Notice that only six data points lay outside the prediction interval.

second one describes the vertical convection currents by means of the temperature and the relative humidity. High temperature and low humidity are associated with intense radiation flux, which occurs mainly in summer months, producing strong vertical mixing (Kasper et al., 1998). The third term due to the horizontal northwest wind is only representative for the α activity which is more sensitive to changes in the origin of the transported aerosols. All these strengths contribute to the dynamics of the particles suspended in the air in a very direct way since the relationship between activity values and each of them is linear. It is outstanding that including the electric field in the models eliminates the need to include any dependence on pressure and precipitation.

To exclude the atmospheric pressure and the precipitation from the fit, we checked that the activities were not too closely associated to any of these meteorological variables by studying the plots of the residuals versus each of them (Hogdin et al., 1982). The patterns obtained are point clouds which suggest that there are no relationships that should have been accounted for in the model. Furthermore, two autoconsistency tests have been carried out. The first one is based on the assumption that if an adequate regression model have been found, there

should be no further significant autocorrelation pattern left in the residual series. The autocorrelation coefficients (r_k) have been computed for k from 1 to 12, as well as its approximate standard error ($s(r_k)$), according to an expression due to Bartlett (Pankratz, 1991). All the obtained values correspond to probabilities lower than 5%, showing that no significant autocorrelation is present. The second test concerns the normality of the residuals. To check that the residuals are normally distributed we have applied the Kolmogorov–Smirnov test finding that the results are consistent with a normal distribution with an associated probability of 0.94 for the α model and a probability of 0.89 for the β one.

Figs. 2 and 3 show the predictions of our models for α and β activities and their associated 95% confidence interval for a period of time comprised from April 1994 to December 1995. The experimental values are also shown. A measurement not included in the confidence interval can be considered as an anomalous value, with a probability of 95%. Each of the regression models describes the normal natural background of α or β emitters, so it will enable us to decide whether a seemingly too high value could have been produced by the meteorological conditions or not. The existence of a model also makes

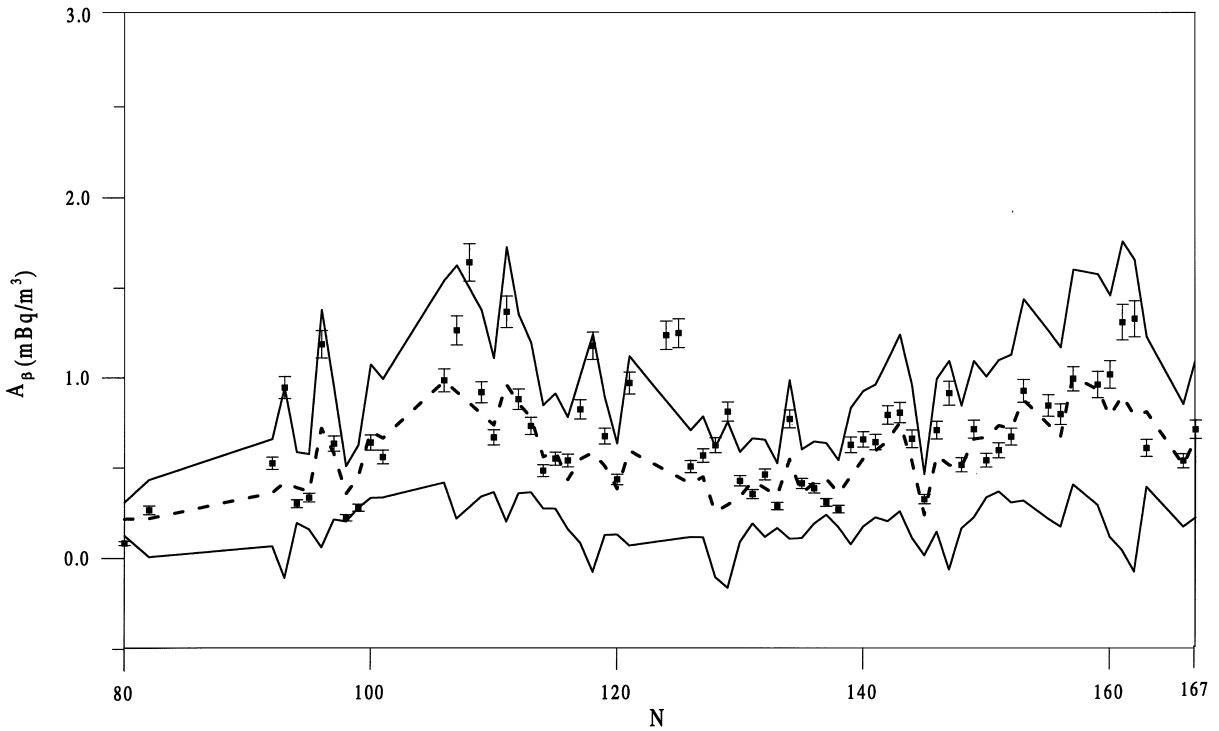


Fig. 3. As in Fig. 2 but for the β activity. Four cases lay outside the confidence interval.

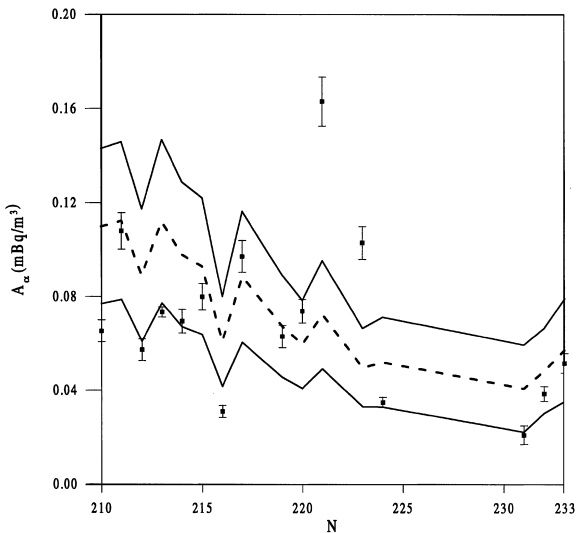


Fig. 4. Activity predictions of the α activity model (average and confidence interval), for data sampled subsequently to the studied years. 75% of the experimental data do not differ from the model predictions.

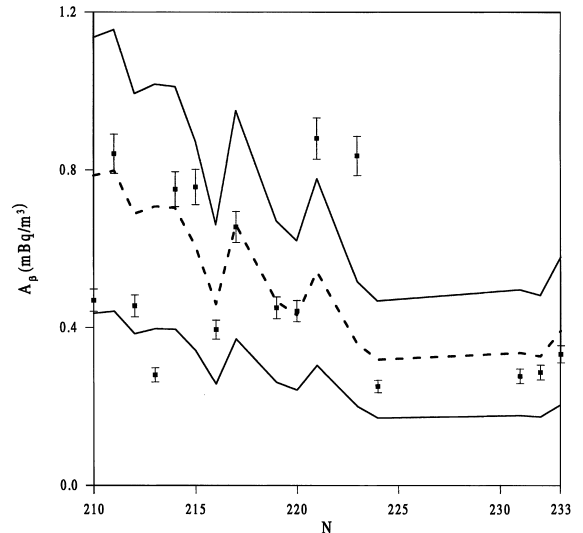


Fig. 5. As in Fig. 4, but for the β activity. In this case 80% of the data is well represented by the model.

evident the presence of data which do not follow the overall tendency and otherwise would have gone unnoticed. These data can indicate the action of

unusual sources. For instance, notice that six experimental points in Fig. 2 and four in Fig. 3 lay out of the confidence interval. For them, the probability of belonging to the distribution given by the model is less than 5%.

Therefore, for these points further investigation is needed because they could indicate the action of an sporadic source of radioactivity or any anomaly in the measurement process.

We have tested both models for different weeks of 1996 and 1997. Only those weeks for which all the meteorological data were available could be considered. The experimental activity data were compared to the predicted values, calculated with the meteorological variables by using the same regression equations that we had obtained for the modeled period. The results are shown in Figs. 4 and 5. We can observe that the models keep on working properly, since the 95% confidence interval comprises most of the experimental activities. This test shows the prediction capability of the model.

4. Conclusions

The concentration of radioactive aerosols in air at Salamanca comes mainly from atmospheric radon descendants. The gross α and β activity levels present a common seasonal trend with maxima in summer, minima in winter, and higher values in autumn than in spring. It has been shown that the seasonal variations can be explained by the relation of the airborne radioactive levels to the meteorological conditions. The atmospheric electric field is the variable most closely related to the activities. But, the influence that both temperature and humidity exerts on the activities is also noticeable. We have proposed a model for the α and β activities based on weather variables. As the statistical tests assure, the better choice of variables are terrestrial electrostatic field, temperature, humidity and NW wind frequency. The simplicity of both models is mostly due to the electrostatic field. Its inclusion enables us to reduce the number of regression variables needed to describe the gross activity variations and improves the prediction capability. The equations reflect the importance in the activity distribution of different strengths affecting the formation, dispersion and deposition of atmospheric aerosols. The electric force is predominant in winter and that is due to the convection currents in summer. This behavior is expected to be valid for other geographical areas. The models also allow to determine whether a given observation can be considered to belong to the usual population or not, and therefore to detect the presence of a hot spot.

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