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Abstract

Gross alpha and gross beta activities were measured weekly in the airborne of the Universitat Politècnica de Valencia campus (in the east of Spain) during the period 2009-2015. The geometric mean values of weekly Aα and Aβ were 5.30·10-5 Bq m-3 and 5.77·10-4 Bq m-3, respectively; with an average ratio Aα/Aβ of 0.097. This study highlights the heterogeneity of gross alpha and gross beta activities depending on the different periods of the year. Data show seasonal variations with the highest activity in summer months and the lowest one in winter months. Several atmospheric factors were considered in order to explain this intra-annual variation (wind speed, temperature, relative humidity, precipitations, dust content and prevailing wind directions). Multiple Linear Regression Analysis were performed in order to obtain information on significant atmospheric factors that affect gross α and gross β variability, which could be useful in identifying meteorological or atmospheric changes that could cause deviations in gross α and gross β activity depending on the seasons considered. Models obtained explain more than 60% of variability for global data, and also for winter and spring-autumn months. However, more research is needed to explain gross α and gross β variability in summer months, because the atmospheric factors considered in the MLR explain less than 35% of variability.

Keywords	gross alpha; gross beta; airborne; MLR; seasons; atmospheric factors
Taxonomy	Environmental Monitoring, Radioactivity in Urban Environment, Natural Radionuclides

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Highlights

- Weekly evolution of gross alpha and gross beta in the airborne during 2009-2015
- Seasonal variations of gross alpha and gross beta activities are observed
- Several atmospheric factors were considered to explain the intra-annual variation
- Multiple Linear Regression Analysis depending on the seasons were proposed
- Models explain more than 60% of variability for global data, winter and spring-autumn

Analysis of the evolution of gross alpha and gross beta activities in airborne samples in Valencia

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Abstract

Gross alpha and gross beta activities were measured weekly in the airborne of the Universitat Politècnica de Valencia campus (in the east of Spain) during the period 2009-2015. The geometric mean values of weekly A_{α} and A_{β} were 5.30·10^{-5} Bq m^{-3} and 5.77·10^{-4} Bq m^{-3}, respectively; with an average ratio A_{α}/A_{β} of 0.097. This study highlights the heterogeneity of gross alpha and gross beta activities depending on the different periods of the year. Data show seasonal variations with the highest activity in summer months and the lowest one in winter months. Several atmospheric factors were considered in order to explain this intra-annual variation (wind speed, temperature, relative humidity, precipitations, dust content and prevailing wind directions). Multiple Linear Regression Analysis were performed in order to obtain information on significant atmospheric factors that affect gross α and gross β variability, which could be useful in identifying meteorological or atmospheric changes that could cause deviations in gross α and gross β activity depending on the seasons considered. Models obtained explain more than 60% of variability for global data, and also for winter and spring-autumn months. However, more research is needed to explain gross α and gross β variability in summer months, because the atmospheric factors considered in the MLR explain less than 35% of variability.

Keywords: gross alpha, gross beta, airborne, MLR, seasons, atmospheric factors

1. Introduction

Radioactivity is present in the air due to cosmic radiation, cosmogenic radionuclides (^{14}C, ^{7}Be, and ^{3}H), and naturally occurring radionuclides (^{40}K and actinium, uranium and thorium series) released or re-suspended from the Earth´s crust to the atmosphere (Akcay et al., 2007; Gordo et al., 2015; Piñedo-García et al., 2015). Anthropogenic radionuclides can also be present in the air in case of nuclear or radioactive accident, and radiological dispersal device (RDD), or “dirty bomb” (EPA, 2009).

Sampling airborne radioactivity is an important tool to fulfil different objectives. Among them, for environmental monitoring, to control atmospheric environmental releases and ensure the environment and public health; for process quality assurance and control in and around nuclear and radioactive facilities; and for emergency preparedness and response, to provide a basis for appropriate actions in case of accident or terrorist attack (Papastefanou et al., 2008). In addition, some radionuclides are also used as atmospheric tracers, such as ^{7}Be and ^{210}Pb, in order to study the air mass behavior, aerosol residence time and other atmospheric characteristics (Baskaran, 2011; Gaffney et al., 2004; Rodas Ceballos et al., 2016).

Gross α and gross β measurement in air is a common method of "screening" in case of emergency with an atmospheric radioactive release (EPA, 2009; Thakur et al., 2011). Moreover, its long-term monitoring also provides information about trends in radionuclide behavior.

In normal situation, gross α and gross β origin is mainly explained due to the presence of long-lived daughters of gaseous ^{222}Rn that are attached to aerosols after cluster formation (Papastefanou et al., 2008). Gross beta activity is mainly due to ^{210}Pb ($T_{1/2}$=22.2 years) and ^{210}Bi ($T_{1/2}$=5.01 days). Huang et al. (2009) explained 61 ± 9% of the gross beta activity due to ^{210}Pb, and the rest contribution could be due to ^{40}K, ^{228}Ra (and its progenies) and other natural or artificial beta emitters. Gross alpha activity is mainly due to ^{210}Po ($T_{1/2}$=138.4 days) contribution (García-Talavera et al., 2001). Concentrations of ^{222}Rn decay products in outdoor air are 0.50 mBq m^{-3} for ^{210}Pb and 0.05 mBq m^{-3} for ^{210}Po (UNSCEAR, 2000), that corresponds to a normal ratio $^{210}Po/^{210}Pb$ of 0.1.

Although the source term of gross alpha and gross beta in airborne is usually constant, seasonal variations of these indexes exist. Several studies try to explain the variation with atmospheric factors and dust content (Arkian et al., 2008; Dueñas et al., 2004, 2001, 1999; García-Talavera et al., 2001; Hernández et al., 2005; Huang et al., 2009; Kitto et al., 2005; Piñero-García et al., 2015). Statistical methods are employed so as to stablish prediction models based on these significant factors. In particular, multiple linear regression (MLR) analysis have been performed with acceptable percentages of explanation for global data. However, no seasonal MLR analysis have been performed before. This approach could help to identify which and how significant atmospheric factors affect the variability of gross α and β index depending on the season. This could help to establish better prediction models.

The Laboratorio de Radiactividad Ambiental of the Universitat Politècnica de València (LRA-UPV) is monitoring the aerosol radioactivity (gross α, gross β, ^{7}Be, ^{131}I, etc.) since 1990, as part of the national monitoring program "Sampling Stations Network" (REM, in Spanish) promoted by the Spanish nuclear regulatory authority, Consejo de Seguridad Nuclear (CSN). The LRA-UPV also participates in intercomparison programs organized by the IAEA, CSN and U.S. Department of Energy (MAPEP program).

The aim of this paper is to study the evolution of gross α and gross β activity in airborne of the city of Valencia over the period 2009-2015 and identify different sources of variability using a seasonal multiple linear regression analysis. The models obtained make it possible to estimate the influence of the atmospheric factors on the gross alpha and gross beta activity, in order to find the most relevant sources of gross α and gross β variability in each season. It should be noted that we focus on the gross α and gross β 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. Study area and sampling

Airborne particulate samples were collected weekly on the campus of the Universitat Politècnica de Valencia from January 2009 to December 2015. 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) 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.

Aerosol samples were collected using Eberlyne G21DX and Saic AVS28A air samplers placed approximately 1 m above ground level. The aerosol particles were retained on a cellulose filter of

$4.2 \cdot 10^{-2}$ m effective diameter and 0.8 µm pore size. The filters were changed weekly and the average volume ranged from 300 to 400 m^3 per week. Each filter was put inside a plastic box and kept in a desiccator until it was measured. Dust content in the filters was determined by weighting the filters before and after exposure under the same laboratory conditions. An average value of 10 mg per week was obtained.

2.2. Gross alpha and gross beta activities measurements

Gross α and gross β activities were measured using a low background gas flow proportional counter, Berthold LB 770-2. Individual samples were placed on stainless steel planchets (5 cm diameter) and measured for 1000 min. Prior to counting, air filters were kept in a desiccator for five days to ensure complete decay of ^{222}Rn short-lived daughter products.

The system was calibrated by counting standard samples prepared in the same geometry as the environmental samples. ^{241}Am standard source and $^{90}Sr/^{90}Y$ standard source, both from *Amersham plc* (UK), were employed to calibrate alpha and beta emitters, respectively. The average counting efficiencies were between 26% to 28% for alpha, and between 40% to 44% for beta. Background of the detector was determined once a week with a clean filter placed on a stainless steel planchet and measured for 1000 min. Average background values were 0.04 and 0.74 counts per minute for alpha and beta, respectively.

Decay correction was not considered because the radioactive species were not identified. Gross α and gross β activities were calculated in Bq m^{-3}, and the limits of detection were $0.43 \cdot 10^{-5}$ Bq m^{-3} for alpha activity and $1.20 \cdot 10^{-5}$ Bq m^{-3} for beta activity (Currie, 1968).

2.3. Atmospheric factors

The atmospheric factors studied in the present work are: precipitations (PP) (in tenths of millimeters), temperature (T) (in tenths of °C), relative humidity (RH) (%), wind speed (WS) (km h^{-1}), prevailing wind direction (WD), and dust content (D) (mg m^{-3}). The meteorological factors were collected by the Universitat Politècnica de Valencia’s weather station, which is also located in the sampling point for gross α and gross β activities.

We selected these variables after taking into account the atmospheric parameters that mainly affect Valencia's weather, with a relatively dry subtropical Mediterranean climate, very mild winters and long hot summers, and considering the variables most frequently used in the literature to study the variability of gross alpha and gross beta activities (Arkian et al., 2008; Dueñas et al., 2004, 1999; García-Talavera et al., 2001; Hernández et al., 2005; Huang et al., 2009).

In this study we considered the mean weekly values of temperature, relative humidity, wind speed, and the total monthly amount of precipitations. The wind direction variable measures the weekly prevailing wind direction classified in: Calm (C) (0°), North (N) (33.75°-2.25°), North-East (NE) (2.25°-6.75°), East (E) (6.75°-11.25°), South-East (SE) (11.25°-15.75°), South (S) (15.75°-20.25°), South-West (SW) (20.25°-24.75°), West (W) (24.75°-29.25°), North-West (NW) (29.25°-33.75°). Dust content was also measured weekly.

2.4. Statistical analysis

2.4.1. Descriptive analysis

A descriptive analysis was applied to study the behaviour of A_α and A_β during the period 2009-2015. The distribution of these activities was characterized and the main descriptive statistics were obtained to analyse the data. Moreover, the ANOVA Tukey's HSD test for multiple comparisons was applied to study the statistical significance of the differences in A_α and A_β over the years and seasons. Finally Spearman correlations between A_α and A_β and the atmospheric factors were computed to evaluate the relation between the behaviour of both activities and these factors by pairs.

2.4.2. Multiple Linear Regression

A multiple linear regression (MLR) analysis was applied in order to identify the meteorological variables that mainly affect the variability of A_α and A_β activities. MLR is a multivariate statistical technique used to examine the relationship between a single dependent variable and a set of independent variables. The main objectives of MLR are explanation and prediction. Explanation examines the regression coefficients, their magnitude, sign and statistical inference, for each independent variable. Prediction involves the extent to which the independent variables can predict the dependent variable (Hair et al., 2010). MLR models are expressed in the following format:

$$Y_t = X_t\beta + \varepsilon_t$$

where Y_t is the predicted value at time t, $X_t = (1, x_{1t}, x_{2t}, \ldots, x_{kt})$ is a vector of k explanatory variables at time $t, \beta = (\beta_0, \beta_1, \ldots, \beta_k)^T$ is the vector of coefficients, and ε_t is a random error term at time t, $t = 1, \ldots, N$. The errors terms should be independent and have a Gaussian distribution.

The explanatory power of the MLR is commonly measured by the R square coefficient defined as follows:

$$R^2 = \left(\frac{\sigma^2_{\hat{Y}_t, Y_t}}{\sigma^2_{\hat{Y}_t}\sigma^2_{Y_t}}\right)100\%$$

where $\sigma^2_{\hat{Y}_t, Y_t}$ is the covariance of the forecast and actual values; $\sigma^2_{\hat{Y}_t}$ and $\sigma^2_{Y_t}$ the variance of the forecast and actual values respectively.

3. Results and discussion

3.1. Temporal evolution of A_α and A_β air concentration

The weekly evolution of A_α and A_β air concentration at the Universitat Politècnica de València from 2009 to 2015 is shown in Fig. 1. In general, the evolution of A_α and A_β is similar over the years and Fig. 1 suggests that it exists a cyclical variability which is repeated every year.

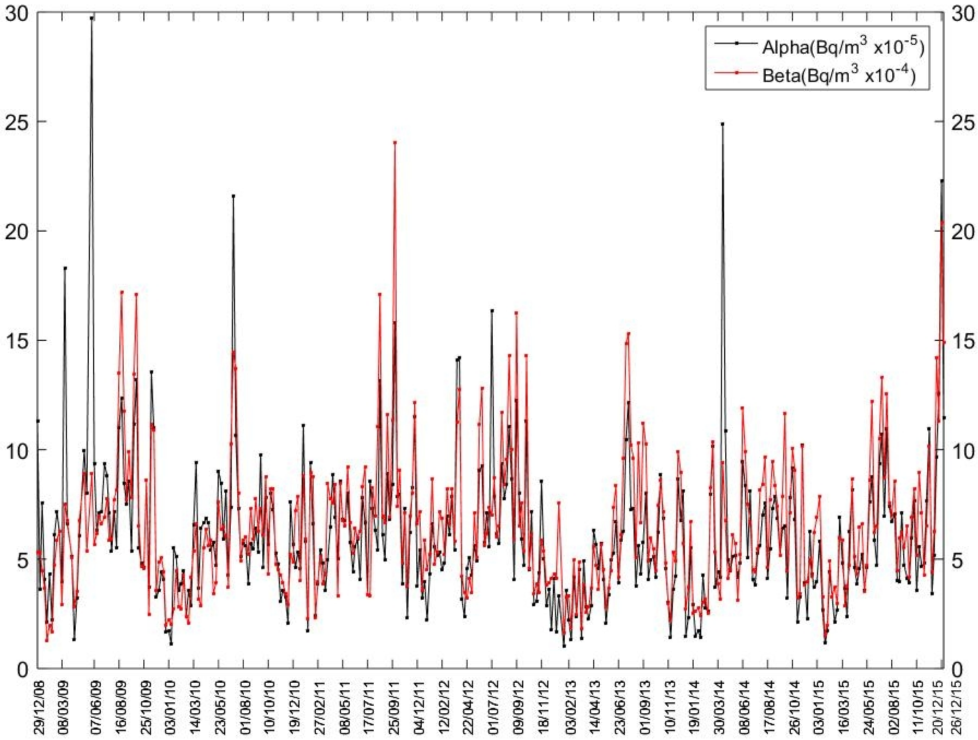


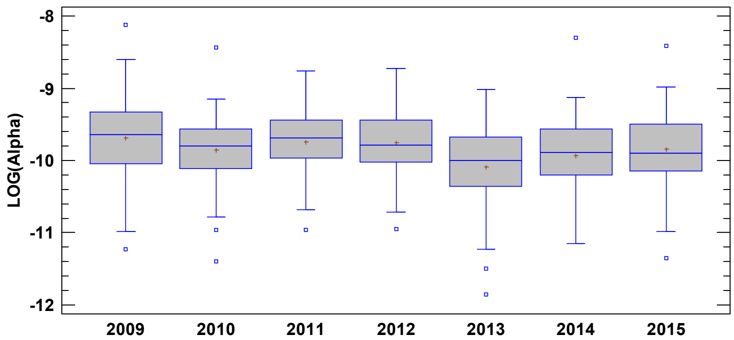
Fig. 1. Temporal evolution of A_α and A_β over the period 2009-2015.

The Kolmogorov-Smirnov test was applied to decide which theoretical distribution fits best to our data. The test confirms that both (A_α and A_β) fits well to a log-normal distribution (p-values obtained were 0.10 and 0.46, respectively). The geometric mean (GM) was therefore used as a descriptive statistic. Descriptive analyses are shown in Table 1. Assuming this log-normal distribution, the GM for A_α was $5.30 \cdot 10^{-5}$ Bq m^{-3}, ranging from $0.71 \cdot 10^{-5}$ to $29.7 \cdot 10^{-5}$ Bq m^{-3}, and the GM for A_β was $5.77 \cdot 10^{-4}$ Bq m^{-3}, ranging from $1.26 \cdot 10^{-4}$ to $24.0 \cdot 10^{-4}$ Bq m^{-3}. Similar results were also presented by Dueñas et al. (2004, 2001, 1999), García-Talavera et al. (2001), Hernández et al. (2005) and Piñero-García et al. (2015). The A_α/A_β ratio was also calculated. In this case the arithmetic mean is close to 0.1, which is the normal ratio for $^{210}Po/^{210}Pb$ (UNSCEAR, 2000). In other studies, different ratios were obtained, such as 0.1 in Málaga (Dueñas et al., 2004), 0.14 in Salamanca (García-Talavera et al., 2001) and 0.2 in Canary Islands (Hernández et al., 2005).

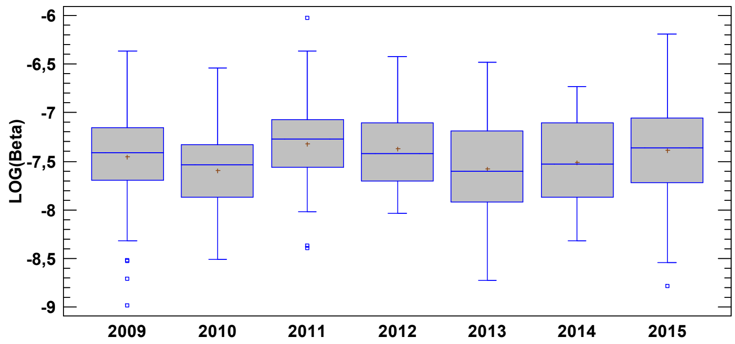
Table 1. Descriptive analysis of gross alpha and gross beta in the airborne of Valencia from 2009 – 2015.

	A_α (Bq m^{-3} ·10^{-5})	A_β (Bq m^{-3} ·10^{-4})	A_α/A_β
Data	361	361	361
Arithmetic mean	6.08	6.48	0.097
Standard deviation	3.42	3.25	0.034
Geometric mean	5.30	5.77	0.092
Geometric standard deviation	1.83	1.64	1.400
Maximum	29.71	24.05	0.334
Minimum	0.71	1.26	0.040

After applying the logarithmic transformation to the activities to assume normality, Fig. 2 shows that the average value and the variability for A_α and A_β are practically constant over the years. The ANOVA Tukey's HSD test confirms this result obtaining a p-value greater than 0.05 for A_β, and only few differences between some years for A_α, which means that there are no significant differences in A_α and A_β over the years. This can be explained because of the principal source term of gross alpha and gross beta in airborne does not change.



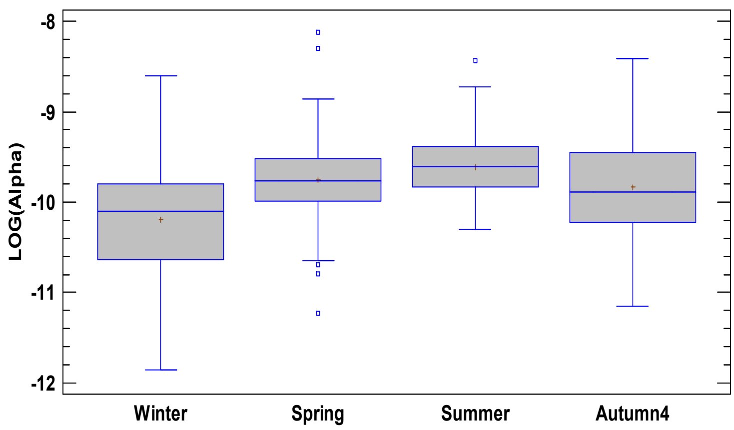
a)



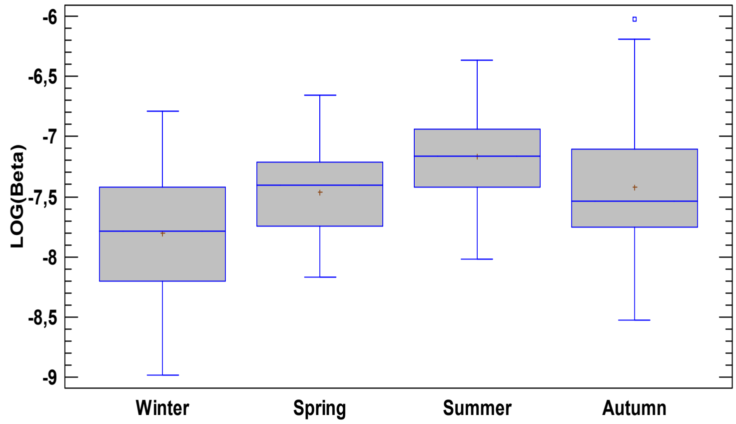
b)

Fig. 2. Box-Whisker plot for the logarithm of: a) A_{α} in Bq m^{-3}·10^{-5} and, b) A_{β} in Bq m^{-3}·10^{-4} over the years.

Fig. 3 shows that it exists an intra-annual variability for both activities with maximum average values in summer, minimum average values in winter and similar average values in spring and autumn. These differences are statistically significant at a 95% confidence level after applying the ANOVA Tukey's HSD test. The main differences are between winter and summer, and spring and autumn could be analysed as a group because they present overlapped ANOVA intervals. The differences observed over the seasons could be mainly explained by the atmospheric factors. Similar results were obtained in other cities of Spain by Dueñas et al. (2001, 1999), García-Talavera et al. (2001) and Piñero-García et al. (2015), with high average activity values in summer and low in winter. The opposite behavior was observed by Huang et al. (2009) in China and Kitto et al. (2005) in New York city, with maximum average gross beta activities in winter months and minimum in summer.



a)



b)

Fig. 3. Box-Whisker plot for the logarithm of: a) A_α in Bq m^{-3}·10^{-5} and, b) A_β in Bq m^{-3}·10^{-4} considering the seasonal factor.

3.2. Evolution of the atmospheric factors

In order to study the influence of the atmospheric factors in gross alpha and gross beta activity in airborne, evolution of temperature, relative humidity, wind speed, amount of precipitations, prevailing wind direction and dust content were analyzed.

Temperature factor has cyclical behavior over the years. Valencia has mild temperatures the whole year, ranging from an average of 25(2)°C in summer and 12(2)°C in winter. The thermal difference during a year is only 13°C.

As a coastal city the relative humidity is very high, with an annual average value of 68(9)%. RH factor also presents an annual cycle, with lower values in winter and higher values in summer than the other seasons. In winter, average RH is 62% with a standard deviation of 10%. In summer, RH presents an average of 74% in summer, and is more constant with only 5% of variability.

Regarding precipitations, Valencia has a dry climate with sporadic rainfall (only 15-20% of rainy days per year). Average total annual precipitations are 318 mm year^{-1}, and they are focused mainly on September and October.

Wind speed maintains constant during the years of the study, with an average value of 5 km h^{-1} (37% RSD). However, this factor presents a cyclical behavior in the year. The average value is practically constant during the year (5 km h^{-1}), but its variability changes with seasons. Relative standard deviation in winter is above 40% with weekly average wind speeds up to 15 km h^{-1}, and 21% in summer months with an average maximum value of 7 km h^{-1}. Winter winds are strong and irregular, while summer winds are soft and regular due to the breeze from the sea.

The analysis of wind direction frequency shows a prevailing absence of wind (Calm) quite constant through the year (average value of 30.7% with 40% of RSD), ranging from an average of 26.3% in winter to an average of 32% in other months. In relation to the other directions, there exist a clear predominance of West and Southwest winds in winter months, with an average frequency of 48%; and East and Northeast winds in summer months, with a total average frequency of 43%. Spring and autumn months show a behavior in between both seasons.

Dust content is another factor considered in the study. Data do not show any tendency, but there are some outliers with higher dust content that can be due to Sahara air masses incoming to Spain or other contributions. The average value is 0.028 mg m^{-3}, which is about 10 mg per filter (in one week of sampling).

3.3. Spearman correlations

Correlations between A_α, A_β and the atmospheric factors considered in the study were calculated in order to evaluate the linear relations between them. A non-parametric technique was used for the analysis because of the lack of normality in gross alpha and gross beta activities. This is a descriptive and individual analysis by pairs of variables. Table 2 and Table 3 show the results obtained.

A strong positive correlation exists between A_α and A_β activity in the airborne of Valencia ($r = 0.85$). This correlation denotes that gross alpha and gross beta origin is the same, and there is no evidence to suggest that there is any anthropogenic contribution to gross alpha or gross beta activities. Other studies observe similar correlations, ranging between 0.72 - 0.88 (Hernández et al., 2007, 2005; Thakur et al. 2011).

Dust content (mg m^{-3}) has also a high linear correlation with both, A_α and A_β. Correlation is positive because of the greater dust content, the higher gross alpha and gross beta activity. However, all dust is not always radioactive and correlation is below 0.5. As can be seen in Table 2, this correlation is more important for alpha than beta.

In Valencia it is found a positive correlation between relative humidity and A_α and A_β (0.60 and 0.55, respectively) like in other coastal cities such as Malaga (Dueñas et al., 1999) and Tenerife (Hernández et al., 2005). However, their linear correlation is very low as compared to Valencia city. On the contrary, cities with continental climate such as Salamanca (García-Talavera et al., 2001) usually present a negative and strong correlation of gross alpha and gross beta with RH. It could be explained because of RH in Valencia is very high and this water vapor in the air benefits cluster formation of radon products.

Correlations with temperature and A_α and A_β are clearly positive as presented in other studies (Dueñas et al., 1999; García-Talavera et al., 2001; Hernández et al., 2005), because of Radon emanates easily from the soil as temperature raises.

Conversely, correlations for PP and A_α and A_β are very low due to the dry climate of Valencia, and negative because rain suppresses Radon emanation from the ground and “washes out” the airborne decay products (Kitto et al., 2005). These results are in accordance with previous studies (Dueñas et al., 1999; García-Talavera et al., 2001; Hernández et al., 2005; Huang et al., 2011).

Correlation of A_α and A_β with wind speed in Valencia was also analyzed. Results show that WS highly influences gross alpha and gross beta activity, but negatively like in other cities (Dueñas et al., 1999; Hernández et al., 2005). This could be explained because wind removes aerosols from air.

Table 2. Spearman correlations between A_α, A_β and the atmospheric factors in Valencia for 2009-2015.

	A_α	A_β	**Dust**	**RH**	**T**	**PP**	**WS**
A_α	1	0.845	0.507	0.606	0.383	-0.183	-0.561
A_β		1	0.348	0.558	0.453	-0.237	-0.586
Dust			1	0.266	0.048	-0.209	-0.268
RH				1	0.410	0.089	-0.523
T					1	-0.138	-0.033
PP						1	0.000
WS							1

In Table 3 correlations between weekly prevailing wind directions and the main other variables are shown. As can be seen, A_α and A_β present positive correlations with Calm, North, Northeast, East and Southeast winds; and negative correlations are obtained for South, Southwest, West and Northwest winds. In particular, East and Northeast (positive) and West and Southwest (negative) present higher correlations because they are the main winds in summer and winter, respectively. Similar results were obtained for correlations between wind directions and, dust content and relative humidity. On the contrary, opposite results are obtained for wind speed and prevailing wind directions because winter winds (W, SW) present higher wind speed than summer winds (E, NE). Moreover, Calm is the absence of wind; therefore, a strong negative correlation is obtained.

As can be concluded from the results, in Valencia winter winds are characterized by high wind speed with mainly W and SW direction, and low relative humidity. In consequence, dust content decreases and also gross alpha and gross beta activity. On the contrary, summer winds are soft and humid with prevailing E and NE direction. For this reason, these more stable atmospheric conditions prevent dust content removal from air and, high relative humidity benefits cluster formation of Radon products. Because of the same origin, the influence of the atmospheric factors is similar for gross alpha and gross beta activity. Other authors also studied this influence. In García-Talavera et al. (2001) studies, significant positive correlation exists with alpha activity and the prevailing wind direction in Salamanca (NW); however, this influence is not considerable on beta activity.

Table 3. Spearman correlations between A_α, A_β, wind speed, dust content and relative humidity with weekly prevailing wind direction in Valencia for 2009-2015.

	C	N	NE	E	SE	S	SW	W	NW
A_α	0.575	0.216	0.431	0.500	0.135	-0.280	-0.524	-0.585	-0.116
A_β	0.592	0.073	0.385	0.511	0.063	-0.303	-0.511	-0.559	-0.151
WS	-0.852	-0.052	-0.128	-0.206	-0.100	0.191	0.355	0.424	0.110
D	0.368	0.219	0.204	0.195	0.224	-0.037	-0.182	-0.353	-0.124
RH	0.551	0.373	0.573	0.518	-0.028	-0.376	-0.608	-0.733	-0.210

3.4. Influence of atmospheric factors in variability of A_α and A_β

Multiple linear regression was applied in order to identify the atmospheric factors that mainly affect the variability of A_α and A_β activities. Two case studies are presented. First, the MLR considers the year as a whole, without distinguishing seasons, called global MLR analysis. Second, the MLR is developed considering the different seasons, i.e. three groups of data are considered belonging to: 1) winter, 2) spring-autumn and 3) summer.

The global MLR analysis (Table 4) shows that 64.3% and 62.8% of the variability of A_α and A_β, respectively, is explained by the atmospheric factors. Lower R^2 were obtained in other studies, such as Dueñas et al. (1999) and Piñero-García et al. (2015) with about 25% of A_α and A_β explanation and 59% of A_β explanation, respectively.

In this study, variability of both activities is not affected by the same atmospheric factors in the global MLR analysis, although A_α and A_β present similar Spearman correlations with them, as we expected (Table 2 and 3). This result is due to the presence of correlation between the atmospheric factors. For instance, RH is positive correlated with A_β (r=0.558) and WS is negative correlated with A_β (r=-0.586). However, RH is not included in the global model of A_β because it explains the same part of A_β variability than WS but in a contrary way (note that RH and WS are strongly negative correlated, r=-0.523). In general, the MLR analysis selects the most relevant atmospheric factors that explain each of them a different part of the A_α and A_β variability, and the analysis excludes, therefore, the atmospheric factors that explain the same as others.

In the global models proposed, A_α and A_β are positively affected by dust content and temperature, and negatively affected by wind speed; same tendencies were obtained in Spearman correlations. However, the influence of relative humidity and wind direction is different in each model. Alpha is affected by RH and SW wind, while variability of beta is explained by SE, N and SW winds. Regarding precipitations, the sporadic rainfall of Valencia makes this factor not significant in gross alpha and gross beta variability.

The meteorological variables are measured in different units of measurement. Therefore, we used the standardized coefficients (the variance of dependent and independent variables are standardized to 1) in order to identify the importance or effect of each meteorological variable on the A_α and A_β variability. These coefficients are shown in Table 8.

Table 4. MLR analysis for global A_α and A_β (361 data).

MLR Model	$\boldsymbol{R^2}$
$LN(A_\alpha) = -10.917(\pm 0.217) + 20.724(\pm 1.966)\mathbf{D} + 0.010(\pm 0.003)\mathbf{RH} + 0.002(\pm 0.0004)\mathbf{T} - 0.091(\pm 0.012)\mathbf{WS} - 1.011(\pm 0.319)\mathbf{SW}$	64.3%
$LN(A_\beta) = -7.430(\pm 0.108) + 10.537(\pm 1.932)\mathbf{D} + 0.003(\pm 0.000)\mathbf{T} - 0.140(\pm 0.010)\mathbf{WS} - 1.692(\pm 0.454)\mathbf{SE} - 1.237(\pm 0.332)\mathbf{N} - 0.949(\pm 0.319)\mathbf{SW}$	62.8%

In addition, considering the different groups of seasons in the analysis (winter, spring-autumn and summer), the meteorological variables could have different influence on A_α and A_β variability.

Therefore, we proposed to apply a MLR analysis for the different groups obtained in the descriptive analysis. Table 5, Table 6 and Table 7 show A_α and A_β models for winter, spring-autumn and summer, respectively. Moreover, Table 8 shows the standardized coefficients of the models.

The atmospheric factors considered explain most of the variability in winter (R^2 > 70%) and spring-autumn months (R^2 ≈ 60%). The main factors are D (positively) and WS (negatively). However, T, RH (both positively) and PP (negatively) also slightly affect A_α and A_β in spring-autumn months. In addition, different wind directions contribute to the models.

However, the influence of the meteorological factors on A_α and A_β variability is much lower in summer months (R^2 < 35%). Gross alpha variability is only slightly explained by RH, and gross beta by RH, T and different wind directions. The evolution of gross α and gross β activity is more constant in summer months, and also the evolution of some of the atmospheric factors, such as wind speed or relative humidity. The low influence of the atmospheric variables could be explained due to these steady-state conditions. Then, part of the A_α and A_β variability could be associated to the uncertainty in the determination of gross alpha and gross beta activities, although, in addition, other factors might be considered in the model trying to explain A_α and A_β variability, such as atmospheric pressure or electrostatic field (García-Talavera et al., 2001). Also alpha and beta activities from previous weeks could be taken into account with a time series forecasting model, which could offer better explanation to the results as proposed by Dueñas et al. (2001). Bas et al. (2017) also employed this method in the evaluation of ^{7}Be air concentration with good results.

Table 5. MLR analysis for winter A_α and A_β (90 data).

MLR Model	$\boldsymbol{R^2}$
$LN(A_\alpha)_W = -9.936(\pm 0.205) + 18.689(\pm 4.414)\mathbf{D} - 0.068(\pm 0.024)\mathbf{WS} - 0.602(\pm 0.325)\boldsymbol{W} - 1.285(\pm 0.615)\boldsymbol{SW}$	71.8%
$LN(A_\beta)_W = -7.445(\pm 0.158) + 8.564(\pm 3.414)\mathbf{D} - 0.120(\pm 0.015)\mathbf{WS} + 1.447(\pm 0.643)\mathbf{E}$	71.3%

Table 6. MLR analysis for spring-autumn A_α and A_β (179 data).

MLR Model	R^2
$LN(A_\alpha)_{SA} = -11.356(\pm 0.342) + 26.057(\pm 2.688)\mathbf{D} + 0.014(\pm 0.004)\mathbf{RH} + 0.002(\pm 0.001)\mathbf{T} + 0.0005(\pm 0.0002)\mathbf{PP} - 0.082(\pm 0.020)\mathbf{WS} - 0.841(\pm 0.411)\mathbf{SW}$	63.7%
$LN(A_\beta)_{SA} = -7.502(\pm 0.151) + 11.138(\pm 2.591)\mathbf{D} + 0.004(\pm 0.001)\mathbf{T} - 0.157(\pm 0.014)\mathbf{WS} - 0.001(\pm 0.0002)\mathbf{PP} - 1.755(\pm 0.616)\mathbf{SE} - 3.975(\pm 1.625)\mathbf{NW}$	56.9%

Table 7. MLR analysis for summer A_α and A_β (93 data).

MLR Model	R^2
$LN(A_\alpha)_{S} = -11.734(\pm 0.490) + 0.029(\pm 0.007)\mathbf{RH}$	17.6%
$LN(A_\beta)_{S} = -10.381(\pm 0.734) + 0.024(\pm 0.007)\mathbf{RH} + 0.007(\pm 0.002)\mathbf{T} + 21.221(\pm 7.263)\mathbf{NW} - 2.929(\pm 0.946)\mathbf{SE} - 0.906(\pm 0.440)\mathbf{NE}$	32.1%

Table 8. Standardized coefficients of MLR models for A_α and A_β (global, winter, spring-autumn and summer).

	MLR Model							
Atmospheric Factor	**Global**		**Winter**		**Spring-Autumn**		**Summer**	
	$LN(A_\alpha)$	$LN(A_\beta)$	$LN(A_\alpha)_W$	$LN(A_\beta)_W$	$LN(A_\alpha)_{SA}$	$LN(A_\beta)_{SA}$	$LN(A_\alpha)_S$	$LN(A_\beta)_S$
D	0.363	0.196	0.356	0.192	0.472	0.229	-	-
RH	0.174	-	-	-	0.241	-	0.419	0.339
T	0.200	0.361	-	-	0.135	0.315	-	0.331
WS	-0.321	-0.522	-0.295	-0.611	-0.240	-0.585	-	-
PP	-	-	-	-	-0.118	-0.152	-	-
C	-	-	-	-	-	-	-	-
N	-	-0.133	-	-	-	-	-	-
NE	-	-	-	-	-	-	-	-0.219
E	-	-	-	0.172	-	-	-	-
SE	-	-0.131	-	-	-	-0.152	-	-0.309
S	-	-0.127	-	-	-	-	-	-
SW	-0.127	-	-0.146	-	-0.119	-	-	-
W	-	-	-0.218	-	-	-	-	-
NW	-	-	-	-	-	-0.128	-	0.282

4. Conclusions

In this research, we have studied the evolution and behaviour of gross alpha and gross beta activity in airborne of Valencia for 7 years (2009-2015), focussing on the descriptive analysis of the data and the influence of the atmospheric factors in both indexes. The results of this work do not show any significant difference in A_α and A_β between years, but there are seasonal variations that we have considered in the study. Spearman correlation between gross alpha and gross beta activity (0.85) show a high linear relation, with a ratio A_α/A_β of 0.1. This result suggests the same origin of alpha and beta mainly due to long-lived radon products. No anthropogenic contribution is observed in the data.

In order to explain seasonal variations of A_α and A_β, the influence of the atmospheric factors was studied. Relative humidity, temperature and dust content increase alpha and beta activity, but precipitations and wind speed remove airborne from the atmosphere. Wind directions were also considered, being prevailing directions West and Southwest winds in winter months, with high speed and variability, and East and Northeast soft winds in summer months with low variability. The former reduces both activities and the latter increases them because of the characteristics of these winds.

Finally, a global Multiple Linear Regression Model was performed to provide information on significant atmospheric factors that affect gross α and gross β variability; and high percentages of explanation were obtained with the factors considered (R^2 > 60%). Moreover, we proposed seasonal MLR models in order to identify atmospheric changes that could cause deviations in gross α and gross β activity depending on the seasons considered. Results show that variability of winter and spring-autumn months is well explained by these factors; however, the explanation in summer months is very low and more research is needed in order to obtain the variables responsible for their variability.

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References

Akcay, S., Tulumen, S., Oymak, S., Kaya, H.I., 2007. Determination of gross α and β activities in Ankara airborne particulate samples in 2003–2004, J. Radioanal. Nucl. Chem., Vol. 273, No.3, 603–608.

Arkian, F., Salahinejad, M., Bidokhti, A.A., Meshkatee, A., 2008. Analysis of gross alpha, gross beta activities and beryllium-7 concentrations in surface air: their variations and statistical prediction model, Environ. Monit. Assess. 140, 325–330.

Bas, M.C., Ortiz, J., Ballesteros, L., Martorell, S., 2017. Evaluation of a multiple linear regression model and SARIMA model in forecasting ^{7}Be air concentrations, Chemosphere 177, 326-333.

Baskaran, M., 2011. Po-210 and Pb-210 as atmospheric tracers and global atmospheric Pb-210 fallout: a Review, J. Environ. Radioact. 102, 500-513.

Currie, L.A., 1968. Limits for qualitative detection and quantitative determination. Application to radiochemistry, Anal. Chem. 40 (3), 586–593.

Dueñas, C., Fernández, M.C., Carretero, J., Liger, E., Cañete, S., 2004. Long-term variation of the concentrations of long-lived Rn descendants and cosmogenic ^{7}Be and determination of the MRT of aerosols, Atmos. Environ. 38, 1291–1301.

Dueñas, C., Fernández, M.C, Carretero, J., Liger, E., Cañete, S., 2001. Gross-α and gross-β activities in airborne particulate samples. Analysis and prediction models, Appl. Radiat. Isotopes 54, 645-654.

Dueñas, C., Fernández, M.C., Liger, E., Carretero, J., 1999. Gross alpha, gross beta activities and 7Be concentrations in surface air: analysis of their variations and prediction model. Atmos. Environ. 33, 3705-3715.

EPA, 2009. Radiological Laboratory Sample Analysis Guide for Incidents of National Significance – Radionuclides in Air, EPA 402- R-09-007, U.S. Environmental Protection Agency Office of Air and Radiation Office of Radiation and Indoor Air National Air and Radiation Environmental Laboratory.

Gaffney, J.S., Marley, N.A., Cunningham, M.M., 2004. Natural radionuclides in fine aerosols in the Pittsburgh area, Atmos. Environ. 38, 3191–3200.

García-Talavera, M., Quintana, B., García-Díez, E., Fernández, F., 2001. Studies on radioactivity in aerosols as a function of meteorological variables in Salamanca (Spain), Atmos. Environ. 35, 221-229.

Gordo, E., Dueñas, C., Fernández, M.C., Liger, E., Cañete, S., 2015. Behavior of ambient concentrations of natural radionuclides ^{7}Be, ^{210}Pb, ^{40}K in the Mediterranean coastal city of Málaga (Spain), Environ. Sci. Pollut. Res. 22, 7653–7664.

Hair, J.F., Black, W.C., Babin, B.J., Anderson, R.E. 2010. Multivariate Data Analysis (7th Edition). New Jersey: Pearson Prentice Hall.

Hernandez, F., Karlsson, L., Hernandez-Armas, J., 2007. Impact of the tropical storm Delta on the gross alpha, gross beta, 90Sr, 210Pb, 7Be, 40K and 137Cs activities measured in atmospheric aerosol and water samples collected in Tenerife (Canary Islands), Atmos. Environ. 41, 4940–4948.

Hernández, F., Hernández-Armas, J., Catalán, A., Fernández-Aldecoa, J.C., Karlsson, L., 2005. Gross alpha, gross beta activities and gamma emitting radionuclides composition of airborne particulate samples in an oceanic island, Atmos. Environ. 39, 4057–4066.

Huang, Y., Tao, Y., Lin, J., Shang-Guan, Z., 2009. Annual cycle of gross β activities in aerosol around Daya Bay area, China, Chemosphere 75, 929–933.

Kitto, M.E., Hartt, G.M., Gillen, E.A., 2005. Airborne activities of gross beta, ^{7}Be, and ^{131}I in New York, J. Radioanal. Nucl. Chem. Vol. 264, No. 2, 387-392.

Papastefanou, C., 2008. Radioactive Aerosols, Volume 12, 1st Edition, Elsevier Science.

Piñero-García, F., Ferro-García, M.A., Chham, E., Cobos-Díaz, M., González-Rodelas, P., 2015. A cluster analysis of back trajectories to study the behaviour of radioactive aerosols in the south-east of Spain, J. Environ. Radioact. 147, 142-152.

Rodas Ceballos, M., Borràs, A., Gomila, E., Estela, J.M., Cerdà, V., Ferrer, L., 2016. Monitoring of 7Be and gross beta in particulate matter of surface air from Mallorca Island, Spain, Chemosphere 152, 481-489.

Thakur, P., Mulholland, G.P., 2011. Monitoring of gross alpha, gross beta and actinides activities in exhaust air released from the waste isolation pilot plant, Appl. Radiat. Isotopes 69, 1307–1312.

UNSCEAR, 2000. United Nations Scientific Committee on the Effects of Ionizing Radiation, Sources and Effects of Ionizing Radiation. United Nations sales publication E.00.IX.3, United Nations, New York.