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

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ABSTRACT

Gross alpha (A_α) and gross beta activities (A_β) were measured weekly in the airborne of the Universitat Politècnica de Valencia campus (in the east of Spain) during the period 2009–2015 (7 years). The geometric mean values of weekly A_α and A_β were $0.53{\cdot}10^{-4}$ Bq m^{-3} and $5.77{\cdot}10^{-4}$ 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.

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 resuspended from the Earth's crust to the atmosphere (Akcay et al., 2007; Gordo et al., 2015; Piñero-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, 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 (Bas et al., 2017a, 2017b, 2016; Baskaran, 2011; Baskaran and Shaw, 2001; Gaffney et al., 2004; Rodas Ceballos et al., 2016; Semertzidou 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 and Mulholland, 2011). Moreover, its long-term monitoring also provides information about trends in radionuclide behavior.

In normal situation, gross α and gross β origin in air is mainly explained due to the presence of long-lived daughters of gaseous ^{222}Rn that are attached to aerosols after cluster formation (Papastefanou, 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). For this reason, the evolution and concentration of ^{210}Pb and ^{210}Po in air and other environmental samples have been deeply studied before (Baskaran, 2011; Persson and Holm, 2011). Their origin is the lower troposphere and global mean 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.

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Although the source term of gross alpha activity (A_α) and gross beta activity (A_β) in airborne is usually constant, seasonal variations of these indexes exist. Several studies try to explain the variation of A_α (or ^{210}Po) and A_β (or ^{210}Pb) with atmospheric factors, dust content and air masses origin (Ali et al., 2011; Arkian et al., 2008; Bourcier et al., 2011; Chham et al., 2017; Dueñas et al., 2011, 2009, 2004, 2001, 1999; García-Talavera et al., 2001; Hernández et al., 2005; Huang et al., 2009; Kitto et al., 2005; Likuku, 2006; Pham et al., 2011; Piñero-García et al., 2015; Tositti et al., 2014; Vecchi et al., 2005). 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 as far to our knowledge. 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 (7 years). Valencia is situated on the east coast of Spain (15 m 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 (Fig. 1).

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% and 28% for alpha, and between 40% and 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. Uncertainties for gross alpha and gross beta activities were approximately 10% and 7% in average, 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 València'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

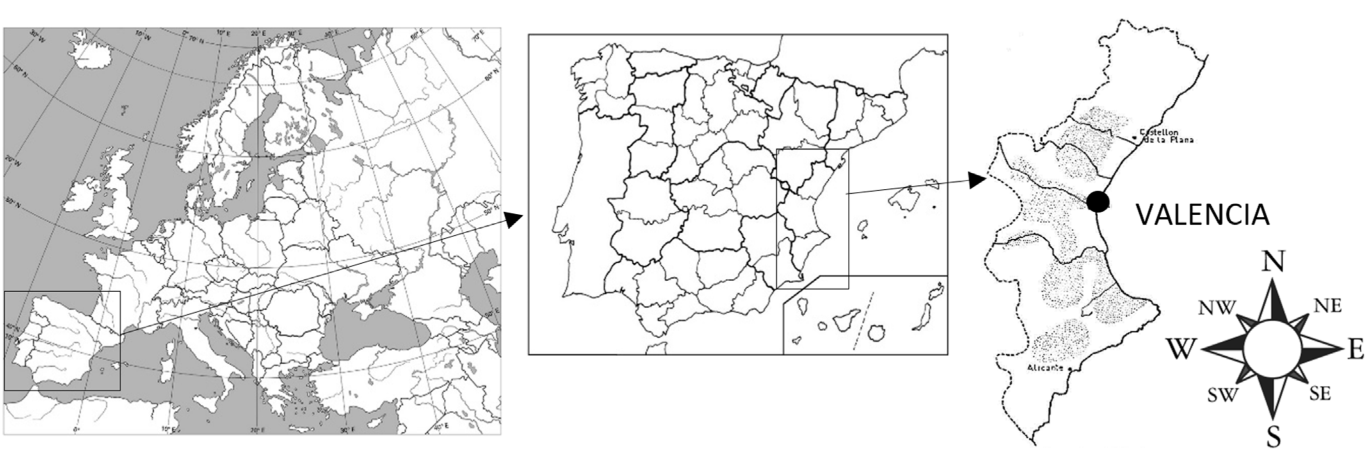

Fig. 1. Map showing the location of Valencia in the center-eastern coast of the Iberian Peninsula.