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Impact of air masses on the distribution of ^{210}Pb in the southeast of Iberian Peninsula air

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ABSTRACT

The current research has been developed in the southern of the Iberian Peninsula in order to better understand the atmospheric processes and also the influence of the air masses origin and pathways in the transport of ^{210}Pb-aerosols in the atmosphere. Simultaneous measurements of the radionuclides ^{7}Be and ^{210}Pb on airborne have been routinely carried out at Granada (Spain 37.177N, 3.598 W, 687 m a.s.l.) from 2010 to 2014. The long term monitoring evolution on ^{210}Pb is discussed in this study and also the useful ratio $^{7}Be/^{210}Pb$.

The maximum monthly activity concentration for ^{210}Pb at ground level in Granada was detected during summer and early autumn (September), whereas minimum activity was measured in the winter. The monthly mean activity concentration for ^{210}Pb was $617.8 \pm 33.0\ \mu Bq \cdot m^{-3}$. The results show that the annual average ^{210}Pb concentrations in samples collected during the same period were almost constant.

The lowest activity concentration for ^{210}Pb are associated with maritime air masses coming from Atlantic and Norwest of Spain, while the highest activity concentrations for this radiotracers were positively correlated with the arrival of mineral dust linked to continental air masses coming from Mediterranean, Africa and Local area. The concentrations values show a nice agreement with the relevant reported results.

1. Introduction

Air pollution by particulates matter (PM) has adverse effects on health, climate and ecosystems, even more when the atmospheric aerosols transport radionuclides (natural and artificial) with the consequent risk associated. Furthermore, considering that the radioactive aerosols have similar behaviour to the aerosols present in the atmosphere, this allows them to be widely used as atmospheric radiotracers. Specifically, the natural radionuclides like ^{7}Be and ^{210}Pb are thoroughly used as radiotracers to better understand the atmospheric processes of the particulates matter (PM) associated to aerosols (Burton and Stewart, 1960; Reiter et al., 1971; Jordan et al., 2003; Papastefanou, 2008; Baskaran, 2011; Lozano et al., 2013) and also their impact in the environment (Bondietti et al., 1987; WMO, 2001; Yutaka, 2013). Moreover, both radiotracers provide useful information regarding the effect of the air masses origin on radioactive aerosols as a result of their different sources, continental and local for ^{210}Pb and cosmogenic for ^{7}Be (which is produced in the lower stratosphere and upper troposphere by cosmic rays). This different origin make them particularly of interest for the study the vertical transport in troposphere, since both radiotracers are transported by convection currents into the troposphere, ascending ^{210}Pb and descending ^{7}Be (Feely et al., 1989) and finally both radionuclides are mainly eliminated by wet removal processes (Liu et al., 2001; Lozano et al., 2012; Heinrich and Pilon, 2013).

^{210}Pb (half-life 22.3 years), is a decay product of the natural ^{222}Rn (half-life 3.8 days, ^{238}U series) by exhalation from the continental soils to the atmosphere (Church and Sarin, 2008; Dueñas et al., 2011). In additions, their concentration could increase due to exogenous sources like burning of coal, use of phosphate

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fertilizers; cars engage exhaust among others (Lozano et al., 2013; Rahim Mohamed et al., 2013). For that reason ^{210}Pb provides information regarding to the influence of the continental air masses on the atmospheric processes.

When radionuclides like ^{210}Pb are released to the atmosphere they are strongly attached to submicron aerosol generating radioactive aerosol (Papastefanou, 2008) which are transported by air masses over the earth's surface and its distribution depends on convection currents in the troposphere, the local climate, pollution and orography of the zone (Heinrich and Jamelot, 2011; Ali et al., 2011). The mean residence times in the troposphere are around 10 days for ^{210}Pb aerosols, while for ^{7}Be is about 20 days, explaining the different origin of their sources respectively. So, the ^{7}Be/^{210}Pb activity ratio is an indicator very interesting for study the vertical transport of air masses in the troposphere (Koch et al., 1996; Arimoto et al., 1999; Ioannidou et al., 2005). Furthermore, the results of Azahra et al. (2004); Dueñas et al., 2009 and Gordo et al., 2015 have demonstrated the seasonal nature of these radionuclides in the surface atmosphere.

To study the effects of the air masses, it is necessary to classify their origins and pathways in order to determine their influences on radioactive aerosols. In this way, clustering analysis (also known as unsupervised classification) has been a usual practice for understanding the Earth's climate and transportation of some particular substances (or tracers) around the world, because it can be applied to find patterns in the atmospheric pressure of polar regions and areas of the ocean that have a significant impact on land climate or for the detection of such tracers. With this idea in mind, and even if sometimes the notion of a well-defined “cluster” (or specific group) is not completely determined, the associated technique of grouping data objects (trajectories in our case) using only information found in the data (spherical coordinates, for example) serves us to classify “similar” trajectories within the same group, different from the other groups where the rest of “unrelated” trajectories will be. In this way, the greater the similarity (measured by some concrete distance: Euclidean, geodesic…) within a group, and the greater difference between clusters, the better the resulting clustering analysis.

The current research has been developed in the southern of the Iberian Peninsula in order to better understand the atmospheric processes and also the influence of the air masses and the local weather conditions on the radioactivity levels of ^{210}Pb measured in aerosols at ground level. So, it is necessary to know the role of air masses origin and pathway about the presence and accumulation of radioactive aerosols in the surface air of the city, since the currents of air masses are the responsible of the transport, concentration and dilution of them and also of its impact on population.

Granada is a natural basin surrounded by mountains of around 1000 m of altitude. The city has a continental Mediterranean climate which is mainly influenced by the orography; isolation of the basin from Mediterranean Sea and also the high altitude of Sierra Nevada. As results, the winter (cold and rainiest season) and summer (low relative humidity and high temperature) are long and extreme seasons while the spring and autumn are a brief transition between both (Piñero-García et al., 2012a,b).

During the cold season, morning thermal inversions are common as a result of long hours of sunshine, few winds, and high traffic flow. Furthermore Granada is influence by frequent Saharan air masses intrusions that transport high concentration of mineral dust, especially during summer time (Guerrero- Rascado et al., 2009). Several research (Hernandez et al., 2005a, 2005b; Dueñas et al., 2011; Brattich et al., 2015) have reported that during Saharan Intrusions events could increase the activity concentration of natural radionuclides like ^{7}Be, ^{210}Pb or ^{40}K in the zone of study. Therefore, this place is a suitable location to study the influence of the air masses coming from African on radioactive aerosols.

For this reason, in our study, deserves special attention the influence of the air masses coming from African continent on radioactive aerosols with ^{210}Pb, since the Saharan Intrusions are the major mineral dust source in this area, about 600–700 106 t/y, increasing also the levels of PM10 in the Western Mediterranean (Rodriguez et al., 2001; Escudero et al., 2011; Prospero, 1999). Since Sahara dust is transported toward European and American continents through the Mediterranean Sea (Querol et al., 2009; Prospero, 1996, 1999), and also it was detected over southern of the Spain by Lyamani et al. (2005).

A full study has been carried out based on the results of different statistical methods that are widely used in environmental science like Pearson Correlation, Cluster Analyses, or Multiple Regression Analysis. Therefore, the whole statistical methods and tests employed in the current research provide powerful information in order to assess the relevant role of this radiotracer and their relationship in the atmospheric processes.

2. Material and methods

2.1. Study place

The atmosphere monitoring station of Radiochemistry and Environmental Radiology Laboratory (LabRadIq) of the University of Granada (UGR) is settled down on the roof of the Faculty of Sciences (UGR) (37.177N, 3.598 W, 687 m a.s.l.). The station is composed by an air sampler pump Radeco AVS-60A. The pump works with a mean flow rate of 85 L min^{-1} sampling an average volume of approximately 800 m^{3} per week. During the current research, ground level atmospheric aerosols were weekly collected, in continuous, on a cellulose filter of 4.2 cm of effective diameter and 0.8 μm pore size from January 2010 to December 2014. After collection, the filters were placed in a Petri dish and then they were stored in desiccators until the measurement to avoid contamination or alteration of the samples. Further details of the sample point and procedure of collection were previously described in the reference Piñero-García et al., 2015.

2.2. Gamma spectrometry

The samples were weekly collected however the radiological characterization was monthly carried out to improve the measurement accuracy of ^{210}Pb. Therefore, the filters belonging to each month (4–5 filters) were analyzed together by gamma spectrometry using a coaxial reverse-type germanium detector with vertical configuration in order to determine the activity concentration of ^{210}Pb and also ^{7}Be.

The detector's efficiency has been calibrated with gamma standards supplied by CIEMAT (Spanish Research Centre for Energy, Environment and Technology). For that purpose, two standards were prepared with the same geometry and composition such as those used for collection of the atmospheric aerosols. One standard with ^{210}Pb and the second one composed by a gamma cocktail (^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{137}Cs, ^{88}Yand ^{60}Co).

Each monthly sample was measured with a counting time of 90 000 s. Then, the spectrums were analyzed to identify ^{7}Be and ^{210}Pb by the photo-peaks generated in the spectrum at the energies of 477.6 keV (Yield 10.42%; Counting Efficiency 3.2%) and 46.5 keV (Yield 4.3%; Counting Efficiency 11.9%), respectively. The activity concentration was corrected to the middle point of the collection period by radioactive decay, considering the half-life of ^{7}Be (53.3 days) and ^{210}Pb (22.3 years).

In addition, the precision and quality of the radiometric measurements and analyses was tested through participation in the