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Mesoscale behavior of ^{7}Be and ^{210}Pb in superficial air along the Gulf of Cadiz (south of Iberian Peninsula)

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HIGHLIGHTS

- Mesoscale analysis of ^{210}Pb and ^{7}Be in air concentrations and bulk deposition.
- High mesoscale correlation between ^{210}Pb and ^{7}Be in coastal area.
- Similar patterns of ^{7}Be and ^{210}Pb in surface air and depositional fluxes.
- Different influence of local winds on ^{210}Pb and ^{7}Be concentrations.

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ABSTRACT

^{210}Pb and ^{7}Be activity concentrations in surface air and in bulk deposition have been measured from November 2009 to December 2011 along the Gulf of Cadiz (Southwest of Spain). This area presents mesoscale variations in its meteorological conditions, with influence of air masses with different origins: maritime, either from the Atlantic Ocean or the Mediterranean Sea and continental, from Iberian Peninsula and north of Africa, which make possible this region to be suitable to analyse the mesoscale spatial and temporal variations of atmospheric compounds. The objective of this study is to determine if there are differences in ^{210}Pb and ^{7}Be activity concentrations in surface air and in bulk deposition at the mesoscale level in this complex area of southwestern Iberian Peninsula taking as reference two sites of the same geographical area but influenced by different meteorological conditions.

The temporal evolution pattern of PM_{10} was different for each site (no correlation between both series was found), but the PM_{10} average concentrations were similar for both locations (differences were not found at 0.05 significance level). On the other hand, the temporal evolution of ^{7}Be and ^{210}Pb activity concentrations in surface air show a good correlation between both sites, indicating this fact a similar behaviour of these radionuclides in the area.

Finally, for each location a strong correlation between ^{210}Pb and ^{7}Be depositional fluxes was also observed, showing that wet deposition plays a key role in the deposition fluxes of both radionuclides. The averages depositional fluxes for ^{7}Be and ^{210}Pb are 750 Bq m^{-2} y^{-1} and 60 Bq m^{-2} y^{-1} in both locations, respectively.

This set of results allows to determine that both radionuclides (^{7}Be and ^{210}Pb) present similar atmospheric behaviours, although with mesoscale variations in the magnitude of the values along the entire southern coast of the Iberian Peninsula.

1. Introduction

Natural radionuclides from terrestrial and upper atmospheric sources (e.g., ^{222}Rn, ^{220}Rn, ^{212}Pb, ^{210}Pb, ^{7}Be, ^{10}Be, etc.) and radionuclides with anthropogenic origins (^{85}Kr, ^{137}Cs, ^{90}Sr, etc.) are widely used as tracers to examine atmospheric processes relevant

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to air quality and climate. Therefore, the long-term measurements of these radionuclides in aerosols are capable of providing useful information on the atmospheric processes (transport, removal and residence time) of aerosol species (Jordan et al., 2003).

Beryllium-7 ($T_{1/2}$ = 53.5 d) is produced throughout the upper troposphere and lower stratosphere as a product of the spallation of oxygen and nitrogen nuclei by energetic cosmic rays (Lal et al., 1958). Because of the short average life span and long residence time of approximately one year of stratospheric aerosols (Kuroda et al., 1962), most of the ^{7}Be nuclei produced in the stratosphere do not readily reach the upper troposphere, except during the spring when a seasonal thinning of the tropopause occurs at mid-latitudes, resulting in air exchange between the stratosphere and the troposphere. In addition, due to its cosmogenic origin the ^{7}Be concentration increases with altitude from the surface of the Earth, and its flux to the Earth's surface shows a latitudinal pattern (Lal and Peters, 1967) that is independent of the geography at any particular latitude (Turerian et al., 1983). Therefore, the standing crop of ^{7}Be in the atmosphere should be the same over the ocean and the continent.

The main source of ^{210}Pb (half-life: 22 years) is the radioactive decay of ^{222}Rn (half-live: 3.8 days) emitted to the atmosphere from the earth's crust; the other artificial sources (burning of coal, use of phosphate fertilizers, cars engage exhaust, fires) in the air have been evaluated as negligible by many authors (Hotzl and Winkler, 1987; Jaworowski et al., 1980). The vertical profiles of ^{222}Rn in the atmosphere show the presence of ^{222}Rn up to the tropopause, which serves as a barrier to the Rn gas, except during strong convective updrafts when some of the ^{222}Rn can reach the lower stratosphere leading to production of ^{210}Pb (Moore et al., 1977). Highest concentrations of ^{222}Rn are most commonly found in the continental boundary layer (3–8 Bq m^{-3}), while it usually decreases by more than one order of magnitude near the tropopause, with levels around 40 mBq m^{-3} (Moore et al., 1977; Liu et al., 1984; Kritz et al., 1993). Large variations in ^{222}Rn concentrations in surface air have been widely reported, depending of the sources of air mass (Church and Sarin, 2008).

However, ^{210}Pb does not necessarily decrease with altitude from the surface, being its distribution with the altitude highly variable and it is commonly as high near the top of the troposphere as near the surface, even sometimes with a reduction at mid-altitudes due to rainout/washout, but this is dependent on the presence of land masses, precipitation, etc. Some authors have found that ^{210}Pb concentrations tend to decrease with altitude in troposphere, measuring the lowest concentrations close to tropopause at 9 km, while in the lower stratosphere the concentrations are little higher (Kownacka, 2002).

Once ^{7}Be and ^{210}Pb have been produced, they are immediately attached to sub-micron-sized aerosol particles (Papastefanou and Ioannidou, 1995). Due to the high reactivity of these radionuclides and their different origins, changes in the ^{7}Be/^{210}Pb activity ratio has been used as indicator to discriminate between both the continental and local sources of aerosols: low values of the ratio (due to high ^{210}Pb levels) reflect a high continental influence, while high ratios indicate a relative isolation from continental sources, showing a maritime influence. Moreover, the temporal and spatial variations in this ratio reflect both vertical and horizontal transport in the atmosphere (Baskaran, 2011).

It is usual that the characterization of the ^{210}Pb and ^{7}Be around the world is performed taking as reference one sampling station that represents the behaviour of both radionuclides in a determinated area (Rulik et al., 2009). However, there is a lack in the knowledge of the local differences in the ^{7}Be and ^{210}Pb behaviour in small areas, which, due to the different origin of both radionuclides, would increase the knowledge about local meteorological conditions.

The Gulf of Cádiz (southwestern Iberian Peninsula) is extended about 320 km from Cape Saint Vincent (Portugal) to Gibraltar, and it is enclosed by the southern Iberian and northern Moroccan margins, west of Gibraltar Strait. This area highlights by the confluence of two completely different meteorological areas: Atlantic and Mediterranean, being also determined the meteorological conditions by the presence of two main orographic elements: the Guadalquivir valley and the Strait of Gibraltar. It is very suitable for the measurement of ^{210}Pb and ^{7}Be in surface air under the influence of air masses with different origins, i.e., maritime (either from the Atlantic Ocean or the Mediterranean Sea) and continental (from Northern Spain and north of Africa). Therefore, the combination of these facts suggests that ^{7}Be and ^{210}Pb could present different behaviour in this area, and makes this place an optimal site for detecting the mesoscale differences in the ^{7}Be and ^{210}Pb behaviour.

Therefore, the aim of this study is to provide a detailed analysis about the mesoscale variations of ^{7}Be and ^{210}Pb concentrations in a complex area of southwestern Iberian Peninsula. With this purpose, the ^{210}Pb and ^{7}Be activity concentrations in surface aerosols and in bulk deposition samples have been determined at two sampling stations (El Carmen and Puerto Real) during a period of about 2.5 years (November 2009–December 2011). The differences have been investigated based on the relationship between activity concentrations in surface air and depositional flux of ^{7}Be and ^{210}Pb in each sampling station; deposition velocities of the aerosols obtained through ^{7}Be and ^{210}Pb concentrations in the air surface aerosols and the total deposition fluxes, and finally, relationship between the ^{7}Be and ^{210}Pb activity concentrations in surface air and depositional fluxes with temperature and rainfall.

2. Materials and methods

2.1. Study area and sampling

The activity of ^{7}Be and ^{210}Pb has been determined in the PM_{10} fractions of air surface aerosols at two different locations in the south-western Iberian Peninsula (Fig. 1); El Carmen (code "EC") station (located at Huelva city) and Puerto Real (located at Cadiz) station (code "PR"). El Carmen is an urban monitoring station located a certain distance (200 m) from the traffic emission sources in Huelva city (37°16′07″ N, 6°55′27″ W) and approximately 3 km from an industrial chemical complex, while Puerto Real is situated in an industrial area close to Cádiz city (36°31′49″N, 6°12′45″W).

The distance between both sampling sites is 100 km and covers the entire coastal area of the Atlantic coast of Spain. However, while Huelva city is located in the surroundings of the valley mouth, Cadiz city is closer to the Gibraltar strait. This different location along the Gulf of Cádiz determines that the meteorological conditions (wind, temperature, relative humidity, precipitation) over both stations are different (Castillo Requena, 1989), and therefore, both stations could be considered sited in different meteorological area. As an example, Fig. 1b displays the wind roses representative of the whole period (2000–2007) in each sampling site, being possible to observe the large differences between both. While in Huelva city dominances the combination of southwesterly–northeasterly (Guadalquivir valley axis) and north-westerly flows, in Cádiz prevalences the arrival of westerly–easterly (Strait of Gibraltar axis). These differences in the wind regime derive in differences in term of temperature, relative humidity as well as in the amount of rainfall.

Measurements of aerosol and deposition samples were registered in both monitoring sites. The aerosol samples (PM_{10}, AMAD < 10 μm) were collected simultaneously in both monitoring stations with Andersen PM_{10} high-volume samplers (flow of 68 m^{3} h^{-1}) mounted with quartz microfiber filters QF Scheicher