Atmospheric Environment 67 (2013) 193–202

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

A decade of ^{7}Be and ^{210}Pb activity in surface aerosols measured over the Western Iberian Peninsula

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HIGHLIGHTS

- A decadal of ^{7}Be and ^{210}Pb activity are analysed over Lisbon Portugal.
- The mean value for entire ^{7}Be time series was 4.0 mBq m^{-3}.
- Higher ^{7}Be activity were found in the years 2001, 2003, 2007 and 2009.
- The year 2001 ws a singular one regarding ^{7}Be activity extreme values.
- ^{7}Be activity show negative correlation sign with relative humidity.

ARTICLE INFO

Article history:
Received 14 June 2012
Received in revised form
15 October 2012
Accepted 29 October 2012

Keywords:
Natural radionuclides
Atmospheric tracers
Gamma spectrometry
Aerosols

ABSTRACT

This work presents the analysis of ^{7}Be and ^{210}Pb continuously measured near Lisbon, Portugal, and their correlation with meteorological parameters, between 2001 and 2010. The values are compared against other similar works over the Iberian Peninsula and Eastern Mediterranean countries. Relative Humidity shows the strongest correlation sign, negative. Higher ^{7}Be activity was found in the years 2001, 2003, 2007 and 2009 being the year 2001 a singular one regarding extremes. The use of ^{7}Be as a stratospheric tracer can be interesting to enhance the radionuclide sample frequency due to the meso to local scale of this phenomenon.

1. Introduction

For international monitoring purposes, due to their health impact effects (e.g. Radon) or because they may act as tracers for atmospheric processes, there is worldwide network for radionuclides measurements in several reservoirs. Two of these networks regard anthropogenic and natural sources: (i) to detect and identify any nuclear test, in the aim of "The Comprehensive Nuclear-Test-Ban Treaty" (URL 1) (ii) the Radon mapping, for human exposure purposes concerning its relation to lung cancer (URL 2; JRC-EU, 2005).

The World Meteorological Organization gives indications for the measurements of radioactive substances, namely Radon-222 (^{222}Rn), Krypton-85 (^{85}Kr), Lead-210 (^{210}Pb) and Beryllium-7 (^{7}Be) as part of the global Atmosphere Watch Program (WMO, 2001). ^{85}Kr is mainly used for detecting nuclear tests and explosions, due to its neglecting natural contribution. Since the ^{210}Pb is a decay product of the ^{222}Rn its origin is the lower troposphere. It is considered as aerosol born particle from the moment it attaches itself to the submicron size aerosol particles. It is an indicator of the atmospheric processes related to removal by deposition, aerosol time residences, stability and vertical movements of air masses (Baskaran, 2011). It may also be used to validate chemical transport models in the aerosol module and also assess the models credibility regarding global and regional atmospheric circulations, specially the continental air masses (WMO, 2001; Baskaran, 2011).

^{210}Pb, together with ^{7}Be, is also used as tracer of atmospheric air masses subsidence from the high troposphere to the low stratosphere region of the atmosphere, which is also an indicator (WMO, 2001), and sometimes the only explanation of high ozone concentrations at surface (San José et al., 2005). This is possible, since, in opposition to the ^{210}Pb, the major source of the ^{7}Be measured at surface is the upper troposphere and the lower stratosphere. Here, it is formed through the reactions between the

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http://dx.doi.org/10.1016/j.atmosenv.2012.10.060

cosmic protons and neutrons with the atmospheric carbon, oxygen and nitrogen nuclei, which make the production rate dependent on the solar and geomagnetic activities (Papandreou Sotiria et al., 2011).

The ^{7}Be has a half-life time of 53.3 days and a mean residence time of around 28 days in the troposphere. As short lived specie its activity concentration is also subject to seasonal variations. It has been used in studies of aerosol particle transport (Hernandéz et al., 2008) due to its association with the sub-micrometric aerosol particles (<1 μm) and also in stratospheric air masses intrusion into the low troposphere as in Monte Cimone, in the Apennines (Bonasoni et al., 2000). Statistical inference of stratospheric air mass intrusion down to the troposphere was found in the work of Elbern et al. (1997) over the German mountain peaks of Zugspitze and Wank.

Measurements of ^{7}Be and ^{210}Pb are scarce and extrapolation of conclusions based on that should not be attempted; however some studies have indicated latitudinal variation of ^{7}Be, seasonal and inter-annual variability. Based on data series data spans from 566 to 1721 weekly measurements, over eight locations, Kulan et al. (2006) shows that the average activity of ^{7}Be, and their spread values, are higher in the southern latitudes (Palermo, Dijon and Prague). These authors have also found the same pattern between ^{7}Be activity and the solar minimum, maximum and average activity, measured at the different latitudes where lower values of the ^{7}Be correspond to the maximum solar activity.

References on measured ^{7}Be at 26 stations in the northern hemisphere may be found in the work of Kulan et al. (2006); review data was also performed by Azahra et al. (2003), Piñero García et al. (2012), focused on southern European countries and also on California (see figure on Results section).

As stated before, since ^{7}Be may act as tracer for atmospheric processes and ^{210}Pb may indicate continental transport of air masses, their concentration will differ with the weather systems that affect the regions where the measurements of the radionuclides are made. In this sense, inter-annual and intra-annual variation may be slightly different from one place to another, and also strongly dependent on the years considered into the time series under analysis.

For instance, over Málaga, ^{7}Be seasonal behaviour was found to be similar in the last decade of the 20th century (sampling period refers to the period between 1992 and 1999) and the first decade of the 21st century (sampling period refers to the period between 2000 and 2009), where higher ^{7}Be activity data was found during Spring and Summer. However, the first period showed higher medians (and also averages) in Spring whereas in the second period higher activity data appeared during Summer (Dueñas et al., 2005, 2009).

The same result was also obtained for Granada by Azahra et al. (2003), with a similar time series length (from 1993 to 2001). However, Pan et al. (2011) show that ^{7}Be activity is lower during Summer and Autumn and higher during Spring time. This variability is also present on the results obtained by several studies regarding ^{7}Be activity and its correlation with meteorological parameters.

Since ^{7}Be is mainly introduced into the low troposphere by vertical transport, the surface concentration of this radionuclide may be enhanced during small time peaks and limited surface areas, at meso to local scales. It is therefore very important to gather all the available information at surface, even if it is not synoptic in time. In the present work, a decadal time series analysis of ^{7}Be and ^{210}Pb measured at the west coast of the Iberian Peninsula, near Lisbon, is presented and correlation with meteorological parameters measured in two meteorological stations attempted.

2. Materials and methods

2.1. Measurement site description

The aerosol particles and the meteorological variables were collected near and inside the Portuguese capital, Lisbon. Lisbon is located in the north shore of the Tagus River, near its mouth to the Atlantic, at 38°43′N of latitude and 9°08′W of longitude. The city is known as the “Seven hills’ city”, with altitudes ranging from 80 to around and 200 m, in some cases descending to the river shore (Baltazar, 2010).

According to the Köppen–Geiger climatic classification, the climatology of the last normal 1981–2010 (provisory) shows that Lisbon has a temperate climate with hot and dry summers (URL 3). Annual average temperatures are around 17.4 °C, confined by averages of maximum and minimum temperatures of 21.3 °C and 13.4 °C, respectively. Regarding precipitation, November and December show monthly average precipitations above 100 mm, whereas July is the driest month with 4.2 mm (URL 4).

Regarding winds, annual mean conditions show predominance of Northern and Northwestern winds. However, winter and summer have different sectors for wind origin, during winter time wind blows from N, NE, and SW, or even W, whereas in summer N and NW occur in 40% of the days. It must be also considered that during summer days, breeze winds due to thermal induced circulations are important and may be superimposed to the anticyclone weather pattern. This local circulation show ascending winds from the river to inland and during the afternoon the reverse is observed. Complex patterns may arise due to the interactions of sea (Atlantic) and the river (Tagus) breezes (Alcoforado et al., 2009).

2.2. Aerosol particles

The atmospheric radioactivity assessment was made by collecting aerosol particles using an aerosol sampling station type ASS-500 (Physik Technic Innovation). The sampling station is equipped with a high volume suction pump, a continuous flux meter and an air volume accumulator. The air was filtered through a 40 × 40 cm Petrianov type FPP-15-1.5 filter with an air mass flow tuned to, around, 800 m^{3} h^{-1}, on average. ^{7}Be and ^{210}Pb activities in the samples were measured by high resolution gamma-ray spectrometry. During the decade presented in this study, the filters undergone two different treatments before activity measurements: a) prior to September 2006 filters were calcinated and measured on a HPGe well-type detector (model Canberra GCW2522) with an active volume of 140 cm^{3}; b) after this date, filters were pressed by using an hydraulic press (MATRA, GmbH) to fit a cylindrical geometry (5 cm diameter and 1 cm thickness) and measured on a broad energy HPGe detector (model Canberra BE5030) with an active volume of 150 cm^{3}. The well-type detector was calibrated in efficiency by using a certified mixed gamma-ray solution (Amersham, QCY48) and a certified ^{210}Pb standard solution, with energy photo-peaks ranging from 46.5 keV (^{210}Pb) to 1836 keV (^{88}Y). The broad energy detector was calibrated in efficiency, for the same energy range, by using a mixed radioactive standard customized in a water-equivalent epoxy resin matrix by Eckert & Ziegler Isotope Products, Inc. Both detectors were placed in a lead shield with copper and tin lining to reduce the natural background radioactivity level. Genie 2000 software was used for data acquisition and analysis.

Most of the time, filters on the sampling station were changed on a weekly basis, except in case of equipment failure and troubleshooting.