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Residence time of tropospheric aerosols in association with radioactive nuclides

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Abstract

The residence time of atmospheric aerosol particles is a function of various removal processes, such as dry deposition by impaction, diffusion, sedimentation and resuspension as well as wet deposition by rain drops (precipitation scavenging). Estimation of the mean-residence time of atmospheric aerosols could be based on measurements of the activities and ratios of activities of cosmic-ray produced radionuclides, such as ^7Be and the radioactive decay products of radon-222 emanated from soil into the atmosphere, such as ^{210}Pb , ^{210}Bi and ^{210}Po . It was found that a mean value of about 8 days could be applied to aerosol particles in the lower atmosphere below precipitation cloud levels as resulted by the application of two different methods, i.e. the ^7Be -associated atmospheric aerosols and the radon decay product aerosols at two different locations, i.e. at Thessaloniki, Greece $40^\circ38'\text{N}$ $22^\circ58'\text{E}$ with dry (precipitation free) climate and at Oak Ridge, Tennessee, USA $35^\circ58'\text{N}$ $84^\circ17'\text{W}$ with high precipitation (wet climate), roughly at similar temperate latitudes, but the first one at East longitude and the other at West longitude, respectively.

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1. Introduction

The residence time of atmospheric aerosol particles in the lower atmosphere assuming that the air in the troposphere is considered as a well-mixed reservoir (closed system) is a function of various removal processes, the most important being:

- (a) dry deposition by impaction, diffusion and sedimentation, and
- (b) wet deposition by rain drops (precipitation scavenging)

as a result of processes occurring both within and below the rain cloud. There could be variations in the removal rates at different continental locations of the globe (Lehmann and Sittkus, 1959), over the oceans (Koch et al., 1996), and at high altitudes of the atmosphere (Martell, 1970; Moore et al., 1973) due to changes in meteorological conditions. There is also a dependence of the tropospheric aerosol residence time on the latitude (Ehhalt, 1973; Balkanski et al., 1993; Koch et al., 1996).

The residence time of atmospheric aerosol particles can be estimated by means of radioactive nuclides as tracers, which become attached to aerosol particles and are removed with them as they are scavenged by precipitation or undergo dry fallout (Warneck, 1988). Several methods have been used for estimating the mean residence time of atmospheric aerosol particles. These include measurements of the activities and ratios of (i) cosmic-ray produced radionuclides, such as ^7Be ($T_{1/2} = 53.3$ days) (Shapiro

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and Forbes-Resha, 1976; Winkler et al., 1998; Yu and Lee, 2002) and (ii) radioactive decay products of radon, ^{222}Rn and thoron, ^{220}Rn which emanate from continental surfaces into the atmosphere, such as ^{210}Pb ($T_{1/2} = 22.3$ years), ^{210}Bi ($T_{1/2} = 5.01$ days) and ^{210}Po ($T_{1/2} = 138.38$ days) (Francis et al., 1970; Poet et al., 1972; Lambert et al., 1982, 1983; Marley et al., 2000; Baskaran and Shaw, 2001). However, there is disagreement between the derived values of the residence times due to various processes, including the fact that they refer to different portions of the atmosphere, e.g. cosmic-ray produced isotopes refer to upper troposphere or lower stratosphere, such as ^7Be , and radon decay products, such as ^{210}Pb , ^{210}Bi , and ^{210}Po that refer to lower troposphere and also due to the existence of different sources for some isotopes, such as ^{210}Po .

This paper is dealing with the residence time of tropospheric aerosols as determined by different methods based on the measurements of the activities of radioactive nuclides which are associated with the atmospheric aerosol particles. It is also an update overview of the existing in the literature data, regarding the residence time of tropospheric aerosols.

2. Experimental methods and techniques

The collection of atmospheric aerosol particles was carried out with high-volume jet air samplers, type TFIA-2 of Staplex having glass-fiber filters type TFAGF

810 of Staplex, highly retentive for particulate material, $20.32\text{ cm} \times 25.40\text{ cm}$ ($8'' \times 10''$) in dimensions and 99.28% collection efficiency for submicron particles as small as $0.3\text{ }\mu\text{m}$ and over. The air-flow rate of these samplers was regulated from $1.7\text{ m}^3\text{ min}^{-1}$ (60 cfm) to $1.92\text{ m}^3\text{ min}^{-1}$ (68 cfm), (average $1.84\text{ m}^3\text{ min}^{-1}$ or 65 cfm). The length of each collection period was 24 h. The size fractionation of atmospheric aerosol particles was carried out with aerosol cascade impactors, type Andersen 2000, as follows: (i) the 1-ACFM design was operated at air-flow rate of 28 l min^{-1} ($1\text{ ft}^3\text{ min}^{-1}$). Its stages had effective cut-off diameters (ECD) of 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 7.0 and $11.0\text{ }\mu\text{m}$, (ii) the low-pressure modification which alters the impactor's operation by increasing the resolution in the submicron region, involved a regulated air-flow rate of 3 l min^{-1} , five low-pressure (114 mmHg that is the absolute pressure downstream of the critical orifice) stages for the submicron region and eight atmospheric pressure stages for separating aerosol particles above $1.4\text{ }\mu\text{m}$. The ECDs of the low-pressure stages were 0.08, 0.11, 0.23, 0.52, and $0.90\text{ }\mu\text{m}$, whereas for the upper stages they were 1.4, 2.0, 3.3, 6.6, 10.5, 15.7, 21.7 and $35.0\text{ }\mu\text{m}$ (iii) the high-volume cascade impactors had a regulated air-flow rate either of about $0.57\text{ m}^3\text{ min}^{-1}$ (20 cfm) or $1.13\text{ m}^3\text{ min}^{-1}$ (40 cfm) and the ECDs were 0.41, 0.73, 1.4, 2.1, 4.2 and $10.2\text{ }\mu\text{m}$ for the 20 cfm configuration or 0.49, 0.95, 1.5, 3.0 and $7.2\text{ }\mu\text{m}$ for the 40 cfm configuration at the standard temperature and pressure atmospheric conditions (25° and 760 mmHg). The stainless steel plates

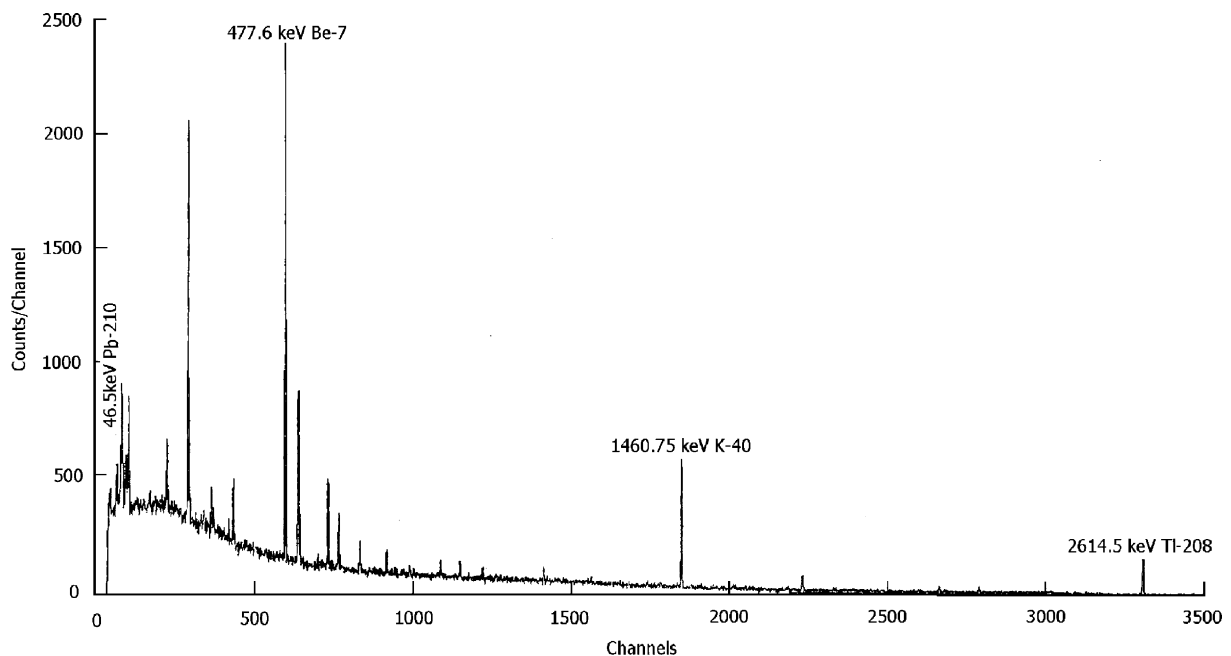


Fig. 1. A typical gamma-ray spectrum of air filter obtained by a Ge detector.

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