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Long-term variations of radionuclides in the Bratislava air

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ABSTRACT

Variations of aerosol radionuclides (2001–2015) in the ground–level air in Bratislava (Slovakia) showed ^{7}Be maxima in spring/early summer and minima in winter, however, an inverse trend was observed for ^{210}Pb, ^{137}Cs and ^{40}K. A decreasing amplitude and splitting of summer maxima for ^{7}Be in the last years has been found. A temporal behavior of the ^{7}Be/^{210}Pb activity ratio showed higher levels during warm seasons due to vertical convection of air masses from higher altitudes. The ^{137}Cs activity concentration in the surface air between 2003 and 2010 was decreasing with an effective half-life of 1.9 ± 0.3 years. The yearly average ^{137}Cs concentrations during 2009–2014 were almost constant, disturbed only by the Fukushima accident in 2011. The increased atmospheric ^{137}Cs and ^{40}K levels observed during the autumn–winter season may be due to surface soil resuspension, biomass burning and radionuclide transport by winds. Seasonal variations of ^{222}Rn activity concentrations were found with maxima at the end of autumn and in winter, and minima in spring. The variability of the average annual course of ^{222}Rn has been larger than that of ^{210}Pb. The ^{210}Pb/^{222}Rn activity ratio was highest at the end of winter and in the spring, while from June to December remained nearly constant. More intensive atmospheric mixing in spring months caused a decrease in the ^{222}Rn activity concentration, while the aerosol component of the atmosphere has been affected mainly during the autumn and winter seasons. The mean residence time of aerosols in the atmosphere was calculated using the ^{210}Pb/^{222}Rn method to be 4.5 ± 0.9 days.

1. Introduction

There have been several sources of radionuclides in the atmosphere, primarily consisting of natural (cosmogenic, primordial and radiogenic) and anthropogenic radionuclides. Cosmogenic radionuclides in the ground-level air are represented mainly by ^{7}Be, which is formed prevailingly in the stratosphere (about 70%) by spallation of light atmospheric nuclei such as nitrogen and oxygen when they absorb protons of the primary component of cosmic rays. The production rate of ^{7}Be can vary with geomagnetic latitude and atmospheric depth (Masarik and Beer, 1999). Variations in galactic cosmic rays (the dominant source of high energy protons in the atmosphere), manifested mainly during 11-yr solar cycles, have also impact on the ^{7}Be production rates, which were discussed by several authors (e.g. Ďurana et al., 1996; Kulan et al., 2006; Leppänen et al., 2010). After its formation, ^{7}Be is almost immediately attached to aerosol particles and follow their transport in the atmosphere. Concentration of ^{7}Be in surface air has been affected by several factors, like air mass exchange between stratosphere and troposphere, vertical transport in the troposphere down to the ground-level air, and horizontal transport to tropics and polar regions (Rosner et al., 1996; Paatero et al., 2001; Ioannidou and Papastefanou, 2006). Typical seasonal variations of ^{7}Be concentrations in the ground-level air have been manifested by spring/early summer maxima. The ^{7}Be has also been used for studying and modeling exchange and transport processes between various layers of the atmosphere (Šimon et al., 2009; Jiwen et al., 2013).

Primordial radionuclides, represented mainly by ^{40}K, ^{238}U and ^{232}Th, are found in most types of soils, and can be easily resuspended into the lower troposphere. They get into the air from surface soil predominantly during spring and autumn when field agricultural activities are frequent, and strong winds transport these radionuclides not only locally but also regionally. Their variations in ground-level air have usually been correlated with other aerosol radionuclides of different origin (^{7}Be, ^{210}Pb, ^{137}Cs), and used as tracers for investigation of surface soil erosion and intrusion of dust particles into the atmosphere (Hötzl and Winkler, 1987; Sýkora et al., 2012; Blazej and Mietelski, 2014). A specific case is

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transport of Saharan sand from North Africa to South Europe (Pham et al., 2005, 2016; Hirose and Povinec, 2015), as well as from Asian deserts in China and Mongolia to East Asia (Korea and Japan) (e.g., Igarashi et al., 2005).

A dominant part of the atmospheric radioactivity consists of ^{222}Rn and ^{220}Rn and their decay products. ^{222}Rn and ^{220}Rn originate in the decay chains of ^{238}U and ^{232}Th, respectively, which are present in terrestrial crust. Gaseous radon enters into the lower atmosphere by emanating and exhaling from soil. The radon behavior in the atmosphere is a result of influence of many factors, e.g. exhalation rates from the ground, solar radiation, changes in the wind velocity and other meteorological factors (e.g. Guedalia et al., 1980; Gesell, 1983; Garzon et al., 1986; Porstendőrfer et al., 1991; Holy et al., 1996, 1998, 2010; Borak and Baynes, 1999; Chambers at al., 2011, 2015). Radon studies in the central European region have not been very frequent. Except the Bratislava station, radon measurements in the atmosphere were carried out in Budapest (Hungary) (Gemesi et al., 1975), in Badgastein (Austria) (Wallner and Ayromlou, 2002), and in Central and South Poland (Podstawczyńska et al., 2010; Zimnoch et al., 2014).

Radiogenic radionuclides which are products of ^{222}Rn and ^{220}Rn decays (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po, ^{210}Pb and ^{210}Po) are valuable natural tracers for the study of atmospheric and soil-atmospheric processes, in particular in the boundary layer near the ground level (Porstendőrfer et al., 1991; Dueñas et al., 1996; Holý et al., 1996; Sesana et al., 2003; Zahorowski et al., 2004; Baskaran, 2011). Atmospheric levels of ^{210}Pb have been of considerable interest as a source of ^{210}Po, which contributes a significant portion of the natural radiation dose to man. There are also other possible sources of ^{210}Pb like traffic or industrial activities. Simultaneous study of ^{222}Rn and ^{210}Pb thus enables to evaluate possible sources of lead from other than natural sources (Fujinami and Esaka, 1987; Gäggeler et al., 1995; Hőtzl and Winkler, 1996; Blaauboer and Smetsers, 1997; Baskaran and Shaw, 2001).

Anthropogenic radionuclides (e.g. ^{3}H, ^{14}C, ^{90}Sr, ^{137}Cs and plutonium isotopes) have been introduced to the atmosphere mainly during atmospheric nuclear weapons testing (known as global fallout) (Livingston and Povinec, 2002; Lujaniene et al., 2009; Povinec et al., 2012a; Hirose and Povinec, 2015), and during nuclear power plant accidents (e.g. Chernobyl and Fukushima) (Povinec et al., 1988, 2012a, b; Hirose and Sugimura, 1990). They have been dispersed all over the world and deposited back to the ground by gravitational settling and precipitation washout. The ^{137}Cs is presently found mainly in soils and vegetation, and it can get back into the atmosphere by soil resuspension (Pham et al., 2011; Povinec et al., 2012a; Sýkora et al., 2012) and by biomass burning (Bourcier et al., 2010; Hirose and Povinec, 2015). Seasonal variations of the ^{137}Cs concentration in the atmosphere, associated with local meteorological conditions (like winds, air temperature, pressure and precipitations) were observed in post-Chernobyl period by several authors (Rosner et al., 1996; Lee et al., 2002; Pham et al., 2011; Bourcier et al., 2011; Povinec et al., 2012a). In the ground-level air, the size distribution of ^{137}Cs carrying aerosols from the post-Chernobyl period was found to be 0.3–1.4 μm (Lujanienė et al., 1997).

All the airborne radionuclides (except those found as noble gases), are attached to fine aerosol particles immediately after their creation, and their behavior is determined by physical and chemical properties of aerosol particles, and by meteorological conditions (e.g., Baskaran, 2011; Pham et al., 2011, 2013; Povinec et al., 2012a; Sýkora et al., 2012; Jiwen et al., 2013; Blazej and Mietelski, 2014). However, our knowledge of aerosol transport in the atmosphere and their removal is rather limited, although these processes are of considerable importance in estimating the hazards following the release of toxic materials in the ground-level air (Ebaid and Khater, 2006).

The aim of this study has been to contribute to better understanding of long-term variations of natural and anthropogenic radionuclides in the ground-level atmosphere. Cosmogenic ^{7}Be, primordial ^{40}K, radiogenic ^{210}Pb and anthropogenic ^{137}Cs have been studied in the Bratislava air since 1977 (Povinec et al., 1988; Beláň et al., 1992; Ďurana et al., 1996; Šimon et al., 2009; Povinec et al., 2012a, b; Sýkora et al., 2012), and investigations of radiogenic ^{222}Rn have been carried out from 1987 (Beláň et al., 1992; Holý et al., 1996, 2010). In the present paper we study radionuclide variations (^{7}Be, ^{40}K, ^{137}Cs, ^{210}Pb and ^{222}Rn) in the Bratislava air for the period from 2001 to 2015. This has been for the first time when such a large set of radionuclide tracers has been simultaneously used for investigation of processes in the ground-level air.

2. Materials and methods

2.1. Sampling

Bratislava, where the atmospheric radionuclides were measured, is an industrial city with 0.5 million of inhabitants. It is situated in the South-western Slovakia near the borders with Hungary and Austria. The climate is continental, warm, mild humid, with cold winters. Average temperatures in January are between −2.5 and −5 °C, and in July between 17 and 18.5 °C. The number of days with rainfall above 1 mm is 92; yearly rainfall is 600–800 mm. The prevailing winds in Bratislava are northwest and northeast winds (Otruba, 1979). Because of the industrial character of the city, Bratislava atmosphere used to be highly polluted. However, since the beginning of the nineties there was a decrease in the air pollution (Povinec et al., 2012a). The samples for radionuclide analysis have been collected on the Comenius University campus (170 m above sea level; 48° 9′ 4″ N; 17° 4′ 14″ E). The campus is situated about 3 km northwest from the downtown, and it is surrounded from all directions by city buildings.

Since 2003, a week periodic sampling of atmospheric aerosols has been carried out, which is a continuation of previous investigations, which started in 1977 with sampling in monthly intervals (Povinec et al., 1988; Beláň et al., 1992; Ďurana et al., 1996). At present, the aerosols are collected using a high volume sampler with flow rate of about 80 m^3 h^{-1} at the height of 2.85 m above the ground. The sampling site is at the Meteorological station of the Faculty of Mathematics, Physics and Informatics of the Comenius University. Nitrocellulose membrane filters (PRAGOPOR 4) with 0.85 μm holes with almost 100% collection efficiency have been used during sampling. The filters are changed once a week, and during each sampling period 11 000–15 000 m^3 of air is pumped through the filters. Corrections for air temperature and atmospheric pressure have been applied to evaluate exact volumes of pumped air through the filters.

2.2. Analyses

Gamma-spectrometry measurements of the exposed filters have been carried out in the Low-background Gamma-spectrometry Laboratory of the Department of Nuclear Physics and Biophysics using two HPGe detectors (Canberra GX 4020 50% relative efficiency, and PGT IGC65-DI 845, 70% relative efficiency), placed in low-level background shields (Povinec et al., 1988; Sýkora et al., 2008) (Fig. 1). Corrections for radioactive decay to the mid-collection period were applied on the measured activity values. The total relative uncertainty of the method for ^{7}Be analysis is about 3%. For ^{137}Cs and ^{40}K activity measurements four one-week filter samples were measured together as a month sample with 5–10% relative uncertainty.

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