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Radioactivity of Bratislava Atmosphere*

I. Sýkora[1], M. Ješkovský[1], J. Merešová[2]

[1]Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University Bratislava, Slovakia
[2]Water Research Institute, Nábrežie arm. gen. L. Svobodu 5, 812 49 Bratislava 1, phone: +421 2 593 43 111, 211

Abstract: The periodic measurements of atmospheric aerosols radioactivity in Bratislava has been started in the Department of Nuclear Physics of the Faculty of Mathematics, Physics and Informatics of Comenius University since the end of the year 2003. The radionuclides concentration of ^{7}Be, ^{210}Pb, ^{137}Cs and ^{40}K is periodically investigated. Average value of cosmogenic ^{7}Be concentration during the observed period was found 2.4 mBq.m ^{3}. ^{210}Pb is long-lived daughter product of ^{222}Rn and its average value of concentration was found 0.8 mBq.m ^{3}. Activity concentrations of ^{7}Be and ^{210}Pb show typical seasonal variations with mutually inverse trends. The activity concentrations were observed to be higher in spring and early summer for ^{7}Be and opposite to that in winter months are higher for ^{210}Pb. The activity concentration of anthropogenic ^{137}Cs in ground level atmosphere is at level 0.6 Bq.m ^{3}. The mean activity concentration of primordial ^{40}K in aerosols reach low values around 5.0 Bq.m ^{3}. The ^{7}Be/^{210}Pb activity ratios are presented and correlation study has been carried out between the meteorological factors and concentrations of radionuclides. The observed mean values can be considered as representative at ground level air in our geographical region.

1. Introduction

The atmosphere represents irreplaceable medium for existence of the human civilization. The exchange processes runing over the atmosphere, as well as the contamination, are the phenomena which can substantilly influence the quality of this medium. It is important to have the possibility to study these processes, to know to suppose their course, possible impact and leakage range of antropogenic pollutants or other substances leaking and spreading in the atmosphere, to have developed high-sensitive methods which are capable to register also small amount of such substances. In the case of radioactive substances, especially anthropogenic radioactive substances, the high-sensitive detection systems and confidential methods of measurements are needed [1, 2]. Anthropogenic radionuclides can leak into the atmosphere during operation, but also at accidents of various nuclear plants. The presence of radionuclides in the atmosphere can be also consequence, for example, of forbidden tests of nuclear weapons. Therefore it is important to develop new methods with threshold of sensitivity as low as possible. The study of atmospheric radioactivity is also one of possible way to investigate the atmospheric processes. Systematic investigation of atmospheric radioactivity is also important from the point of view of the health protection.

Most of the airborne radionuclides, cosmogenic or radon derived, attach to fine aerosol particles immediately after their creation. Hence, the behavior of airborne radionuclides is determined by physical and chemical properties of the aerosol particles to

*) *Dedicated to Associated Professor Martin Chudý on the occasion of his 70th birthday.*

which they are attached and by the actual meteorological conditions. For this reason, the radioactive aerosols may provide useful information on atmospheric thermodynamic processes such as transport and mixing of air masses, dispersion or removal of aerosol particles.

^{7}Be is naturally occurring radionuclide (half-life 53.5 days) of cosmogenic origin formed by spallation of light atmospheric nuclei such as carbon, nitrogen and oxygen when they absorb protons of the primary component of cosmic rays. The highest production rate is at height 15–20 km and declines approximately exponentially with decreasing altitude. Around 70% of the ^{7}Be is produced in the stratosphere and the remaining 30% in the troposphere [3]. ^{7}Be attaches to particles with diameter between 0.7 and 1.1 m [4]. The residence time of ^{7}Be in the stratosphere is around one year and it is about 3–5 weeks in the troposphere [5]. Concentrations of ^{7}Be in low-level atmosphere vary on the level of mBq.m ^{3}. Variations of the annual mean concentrations are connected with changes in the production rate, which depends on the solar activity cycle. Cosmogenic radionuclide ^{7}Be is used as the tracer of stratospheric air masses and in studies of atmospheric transport processes and circulation. Considering that ^{7}Be has pure outdoor origin, it is used as the tracer in experiments examining the ingress of aerosols into buildings. Its seasonal variations appear to show the effect of four factors:
(a) the rate of exchange between the stratosphere and the troposphere, (b) the rate of vertical mixing within the troposphere, (c) transport of air masses from middle latitudes into the high latitudes and (d) the amount of rainfall.

^{210}Pb is a long-lived decay product (half-life 22 years) of uranium series (^{238}U). It gets into the air from radioactive noble gas ^{222}Rn exhaled from the Earth's crust. The ^{222}Rn exhalation rate depends on the soil type and its ^{226}Ra concentration. The variations on the exhalation rate are caused by precipitation and soil moisture. The variations of ^{222}Rn and subsequently of its decay products in air concentrations are induced mainly by atmospheric mixing. Atmospheric level of ^{210}Pb is of a considerable interest like a source of ^{210}Po which contributes a significant portion of the natural radiation dose to man.

Artificial radionuclide ^{137}Cs (half-life 30 years) in the air comes from two main sources. Part of its activity comes from the stratosphere as the remains of the atmospheric nuclear weapon tests. There is also some of ^{137}Cs originating from the Chernobyl accident in resuspended fine soil particles. ^{40}K is classified as stellar or primordial radionuclide (half-life 1.3 10^{9} years), produced in stellar nucleosynthesis. ^{40}K is abundant in the Earth's crust and it gets into the air gets via resuspension from the surface soil.

The aim of our environmental radioactivity monitoring is to study the transport processes in between the troposphere and the low-level atmosphere. Moreover, these aerosol measurements can serve as air monitoring, sensitive to accidents of the nuclear relevance.

2. Experimental

Since the end of 2003 a monitoring of atmospheric aerosols radioactivity has been started with a one week period in the Department of Nuclear Physics at Comenius University in Bratislava [8]. These measurements are the continuation of the aerosols radioactivity investigation realized in this laboratory during the period 1981–1995 in month intervals. The atmospheric radioactivity study recovery started otherwise in 2001 but these measurements did not have full periodic character.

At present, the aerosols are collected using high volume sampler with the flow rate around 80 m^3/h at the height of 2.85 m above the ground. The sampling site is located at the meteorological station close to our university. The special nitrocellulose membrane filters (PRAGOPOR 4) with 0.85 m holes with the approximate 100% collection efficiency are used. The filters are changed once a week and during each sample period about 11 000 m^3 of air are pumped. Corrections for the air temperature and atmospheric pressure are applied to evaluate true volumes of pumped air.

Radioactivity measurement of the exposed filters is performed by standard gamma-spectrometry with two HPGe detectors (EG&G Ortec 51370/20-P with Be window and PGT IGC65-DI 845 with relative efficiency 69%) placed in low-level background shields. Corrections for the radioactive decay to the mid-collection period were applied on the values. The total relative uncertainty of the method is about 3%. For ^{137}Cs and ^{40}K activity concentration measurement 4 one-week samples are accumulated into a month sample and determined with 10% relative uncertainty.

3. Results and discussion

3.1. The activity concentration and seasonal trend

The activity concentrations of the ^{7}Be and ^{210}Pb in Bratislava atmosphere are presented in Figs. 1 and 2. The observed range and average values (Table 1) can be considered as representative at ground level air in our geographical region. The results presented in this work are generally in agreement with the data reported in literature [10–16]. The activity concentrations of ^{7}Be and ^{210}Pb exhibit a pattern of seasonal variations with mutually inverse trends. In our region the ^{7}Be concentrations are higher in spring compared to autumn and winter season. It is connected to the tropopause thinning at mid-latitudes resulting in air exchange between the stratosphere and troposphere. The concentrations of ^{210}Pb reach higher values in autumn and winter months, which is attributed to frequent inversion conditions of the surface air layers. The decrease of the ^{210}Pb concentration in warm spring and summer season is a result of intensive air mixing.

Table 1. Detection limits, range and average values of radioactivity concentrations in Bratislava during the period December 2003 – January 2007 (for ^{7}Be and ^{210}Pb till December 2008).

	Detection limit	Range	Average
^{7}Be	1.2 Bq.m ^{3}	(0.2 5.1) mBq.m ^{3}	2.4 mBq.m ^{3}
^{210}Pb	0.5 Bq.m ^{3}	(0.1 2.4) mBq.m ^{3}	0.8 mBq.m ^{3}
^{137}Cs	0.4 Bq.m ^{3}	(0.4 1.2) Bq.m ^{3}	0.6 Bq.m ^{3}
^{40}K	0.3 Bq.m ^{3}	(1.3 11.8) Bq.m ^{3}	5.0 Bq.m ^{3}

The average concentrations of ^{137}Cs (Fig. 3) and ^{40}K (Fig. 4) are very low and reach the level of 0.6 Bq.m ^{3} and 5.0 Bq.m ^{3}, respectively. In the past, there has been correlation

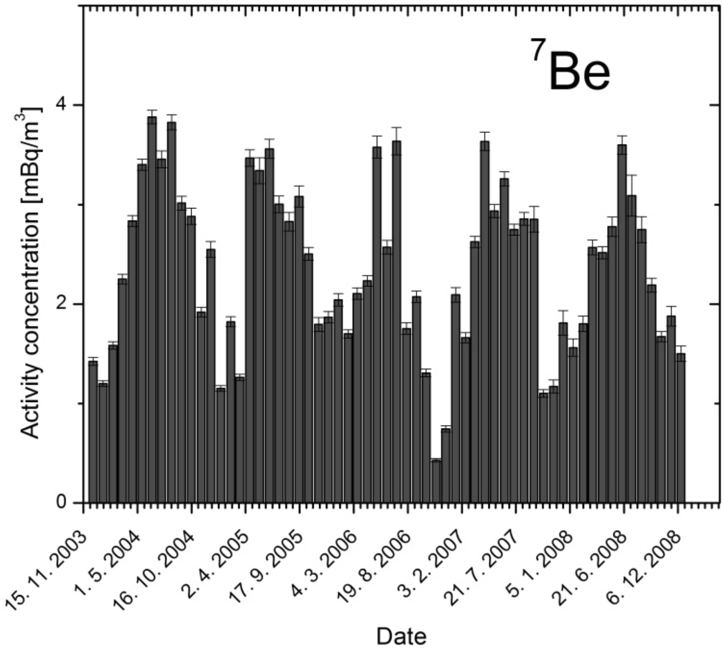

Fig. 1. Temporal variations of the ^{7}Be activity concentration in Bratislava low-level air.

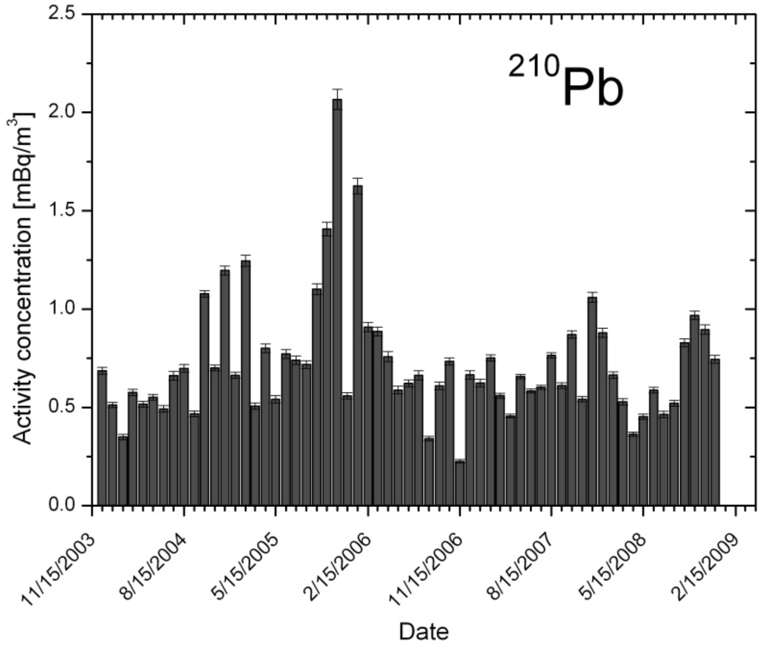

Fig. 2. Temporal variations of the ^{210}Pb activity concentration in Bratislava low-level air.

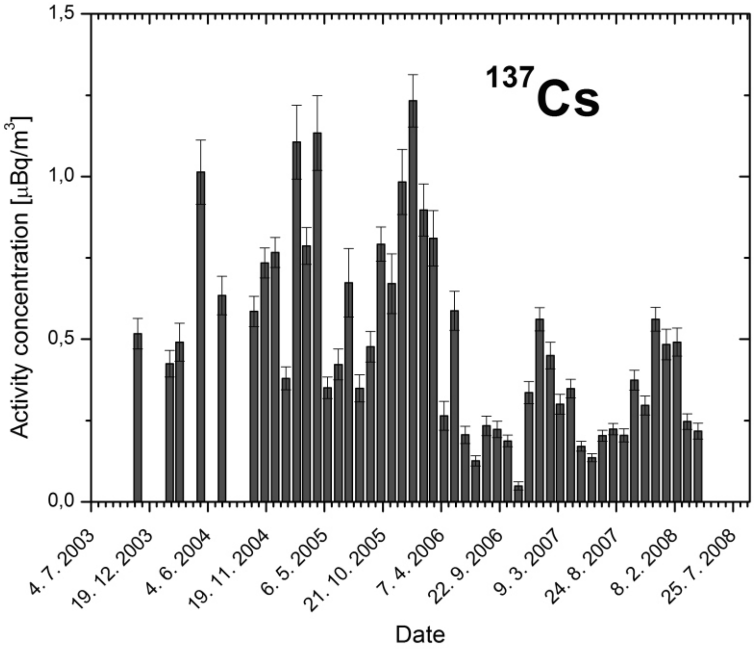

Fig. 3. Temporal variations of the ^{137}Cs activity concentration in Bratislava low-level air.

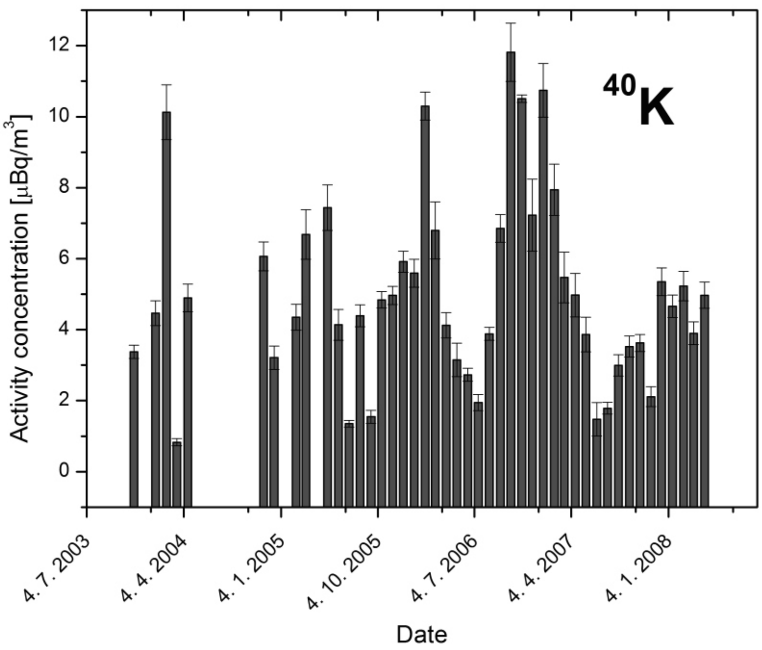

Fig. 4. Temporal variations of the ^{40}K activity concentration in Bratislava low-level air.

between ^{7}Be and ^{137}Cs concentrations observed. The cause was the stratosphere as the reservoir of both radionuclides: the cosmogenic ^{7}Be produced by spallation of light nuclei atmospheric with cosmic rays, and the ^{137}Cs originating from nuclear weapon tests. The present very low concentration of ^{137}Cs is the reason of no correlation found. The potential seasonal variations are ranging within the uncertainty of determination.

3.2. Analysis of meteorological factors

The correlation study has been carried out between some meteorological factors and concentrations of radionuclides. The linear correlation coefficients are listed in Table 2. The only significant correlation was found between the ^{7}Be concentration and air temperature. Weak negative correlations are observed for ^{7}Be with humidity and cloudiness. The one week measurement duration is quite long compared with the typical time scale of variations of meteorological parameters so that the correlation might be slightly better.

Table 2. Linear correlation coefficients between activity concentrations of ^{7}Be and ^{210}Pb and selected meteorological parameters.

Radionuclide	Temperature	Pressure	Humidity	Precipitation	Cloudiness
^{7}Be	0.70	0.17	0.55	0.18	0.44
^{210}Pb	0.25	0.32	0.35	0.20	0.05

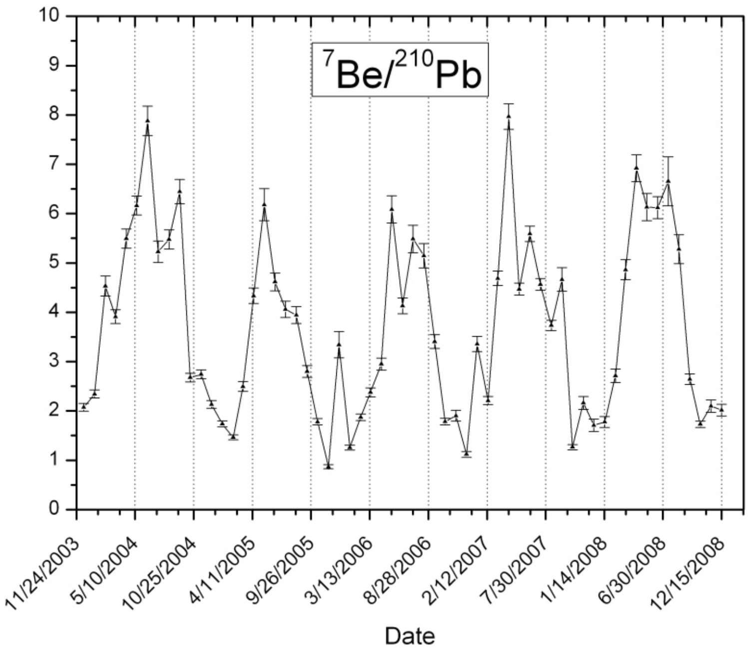

Fig. 5. Temporal variations of $^{7}Be/^{210}Pb$ concentration ratio.

3.3. ^{7}Be/^{210}Pb concentration ratio

Ratio of ^{7}Be and ^{210}Pb, due to their different origin, should depend on the altitude from which the air was transported, on continental influences and on removal processes. In this way the ratio serves as the parameter of the air masses transport history [15]. The values of ^{7}Be/ ^{210}Pb concentration ratio varied from 0.98 to 7.88 exhibit the summer maximums and winter minimums as shown in Fig. 5. It is caused by higher intensity of vertical convection of air in the summer season bringing air rich for cosmogenic nuclides from upper layers and hereby removing the elements of the terrestrial origin.

4. Conclusions

Since December 2003 a continuous weekly monitoring of ^{7}Be and ^{210}Pb concentrations in the low-level atmosphere has been carried out in Bratislava (Slovakia). In a month period the concentration of ^{137}Cs and ^{40}K in atmospheric aerosol has been measured. The observed average values in this paper are in good agreement with the data reported in literature for similar continental locations. The typical pattern of seasonal variations was observed for ^{7}Be and ^{210}Pb activity concentrations. The pronounced maxima in spring/ summer for ^{7}Be and inverse trend for ^{210}Pb with the highest values in winter were measured. The correlation between ^{7}Be concentration and air temperature was found. The temporal behavior of ^{7}Be / ^{210}Pb concentration ratio shows that higher intensity of vertical convection of air in warm seasons which brings the air-masses from higher altitudes.

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