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Temporal Variations of ^{7}Be Activity Concentrations in Slovakia

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Abstract: The temporal variations of concentration of cosmogenic radionuclide ^{7}Be in the atmosphere in Slovakia in time period 1981 2005 are presented in this work. The measurements were performed in two intervals 1981 1994 and 2001 2005 in two localities of Bratislava and obtained results were checked for correlation with solar activity cycle. Activity concentrations of ^{7}Be show seasonal variations with obvious one or two maxima in spring to fall period and a minimum in winter. The mean of ^{7}Be concentration in low level atmosphere is (2.43 ± 0.10) mBq.m ^{3} for the time period 2001 2005. This value is comparable to other locations in Slovakia and other countries.

1. Introduction

The radionuclide ^{7}Be (half-life 53.5 days) is formed by cosmic ray spallation of nitrogen and oxygen in the atmosphere. 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 remaining 30 % in the troposphere [1]. Newly produced atoms diffuse in the atmosphere until they encounter aerosol particles to which they attach. ^{7}Be attaches to fine particles with diameters between 0.7 and 1.1 m [2]. The residence time of ^{7}Be in the stratosphere is around one year and in the troposphere is about 3 5 weeks [3].

The atmospheric aerosols are submicron particles, on which the elements of solid or liquid state present in the air are attached. The aerodynamic size of atmospheric aerosol determined the degree of biological effect on human organism. They are obviously sorted according their size into: (a) the Aitken nuclei mode (from 0.003 to 0.07 m, average 0.015 m), (b) the accumulation mode (from 0.07 to 2 m, average 0.3 m) and (c) the coarse mode (from 2 to 36 m, average >10 m).

Cosmogenic radionuclide ^{7}Be is used as 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 a tracer in experiments examining ingress of aerosols into buildings [4]. Its seasonal variations appear to show the effect of four factors [5]:

(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.

Concentrations of ^{7}Be in low-level atmosphere vary on the level mBq.m ^{3}. Variations of annual mean concentrations are connected with changes in the production rate, which depends on the solar activity cycle.

Nowadays radioactivity of atmosphere is continuously monitored in many places all over the world [6–11]. Also in Slovakia, at the Faculty of Mathematics, Physics and Informatics, there is a long tradition of measuring the radioactivity of the low-level atmosphere [12–14].

2. Experimental and Methods

In the time period 1981–1994 samples were collected at the Hydrometeorological Institute in Bratislava – Koliba [12]. Aerosol particles were captured using the commercial filter (SIXPOR3 – the collection efficiency almost 10 %) with a very dense structure able to capture low-dispersive aerosols. The filters were exposed for up to one month and the total volume of pumped air was measured. After exposition the activity concentration of ^{7}Be was measured by analysis of 477.6 keV gamma-rays using Ge(Li) detector with passive and active shielding. The calculated activity was corrected for the radioactive decay during the sampling time and during the time between the end of collecting and the measurement.

Since the year 2001 there is the sampling location at the Meteorological Station near the Faculty of Mathematics, Physics and Informatics, Comenius University, Bratislava. At height of 2.85 m above the ground aerosol particles are collected on the nitro-cellulose membrane filters (PRAGOPOR – the size of pores 0.85 m, the collection efficiency approximately 100 %) using the sampler device with an air-flow rate of 30 $m^3.h^{-1}$. The filters have been changed every week and during each sample period about 3000 m^3 of air were filtered. In September 2004 the new sampling device was launched and the volume of pumped air increased two times. Corrections for temperature and the reduced pressure were made to evaluate right volumes of pumped air. After exposing in the sampler device the filters have been measured by standard gammaspectrometry with a semiconductor HPGe detector with a beryllium window placed in the low-background shield. The ^{7}Be activity concentrations were calculated by the detection of emitted 477.6 keV gamma-rays. Corrections for radioactive decay to the mid-collection period were applied on the values. The total relative error of the method is bellow 5 %.

3. Results and Discussion

Temporal variations of monthly activity concentration of ^{7}Be in low-level atmosphere of Bratislava in time period 1981–2005 are presented in Fig. 1. In the period 1981–1994, 151 monthly samples were collected. The concentrations of ^{7}Be ranged from 0.70 mBq.m^{-3} in December 1990 to 8.69 mBq.m^{-3} in July 1994, with the mean (3.15 ± 1.22) mBq.m^{-3}. During monitoring period from March 2001 to April 2005, 123 sets of aerosol filters were sampled and the concentrations of ^{7}Be varied between 0.59 mBq.m^{-3} in April 2001 and 5.28 mBq.m^{-3} in April 2005 with the mean value (2.43 ± 0.10) mBq.m^{-3}.

Fig. 2 shows the distribution histogram of monthly activity concentrations of ^{7}Be for the monitored period 1981–2005. Frequency curve was fitted with the Gaussian function. The correlation coefficient is 0.977 and the centre of the peak is (2.84 ± 0.05) mBq.m^{-3}.

Comparing the average values for the two sampling periods, the one from 1981–1994 is higher. This difference is related to changes of the production rate during the time. In Fig. 4 the annual average activity concentrations of ^{7}Be and sunspot numbers for the investigated

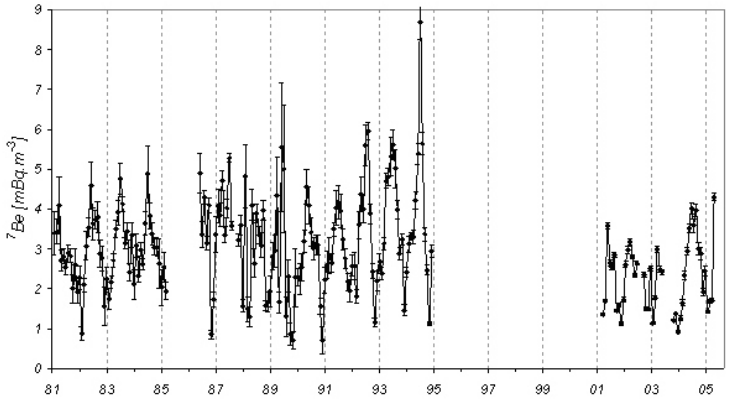

Fig. 1. Temporal variations of activity concentrations of ^{7}Be in low-level atmosphere in Bratislava for the period 1981–2005.

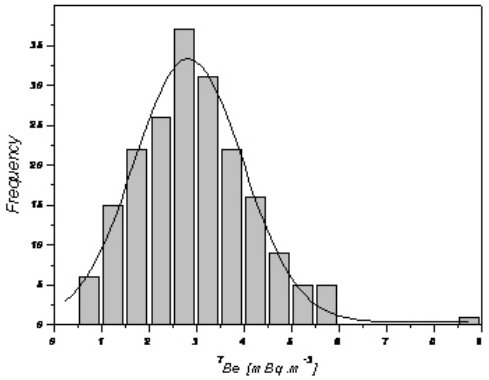

Fig. 2. Frequency distribution of monthly activity concentrations of ^{7}Be for the period 1981–2005. Fitted with the Gaussian function with correlation coefficient = 0.977. The centre of the peak is (2.84 ± 0.05) Bq.m ^{3}.

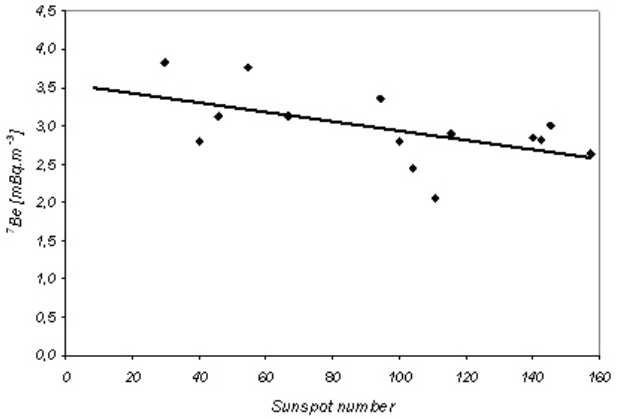

Fig. 3. Correlation relationship between annual averages of activity concentrations of ^{7}Be and sunspot number for the period 1981–2004.

period are presented. The flux of primary galactic cosmic rays in the Earth's atmosphere decreases in the years corresponding to a high sunspot number, whereas it increases in the years when the Sun is relatively calm. Owing to this inverse correlation the production rate of cosmogenic radionuclides decreases with increasing sun activity and vice versa. The period 1981–1994 covers the end of 21st solar cycle and almost whole 22nd solar cycle. But the sampling period since 2001 begins in the tail part of the last solar cycle 23 and the increase of the annual average activity concentration of ^{7}Be in the next years is expected.

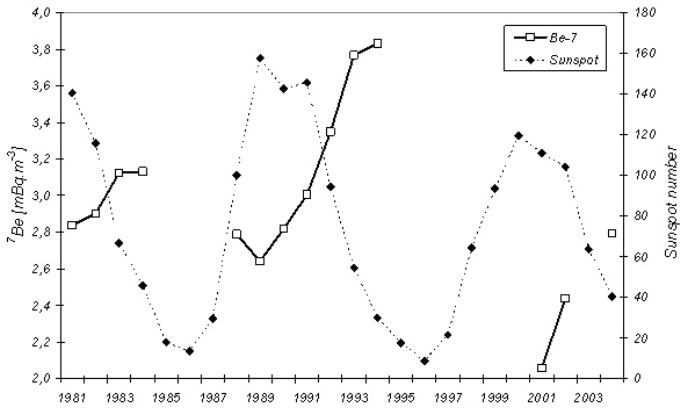

Fig. 4. Annual averages of activity concentrations of ^{7}Be in low level atmosphere in Bratislava and sunspot number for the period 1981–2004.

The seasonal variations of ^{7}Be activity concentrations are analysed by seasonal indices, defined as ratios of the mean monthly activity concentration and the overall average concentration in the monitored period. Seasonal indices for the two sampling periods are presented in Fig. 5. The highest values of activity are observed during spring and summer terms. It is an effect of air-mass transport from stratosphere to troposphere induced by heating of the Earth's surface. The rising temperature enhances the exchange of matter be-

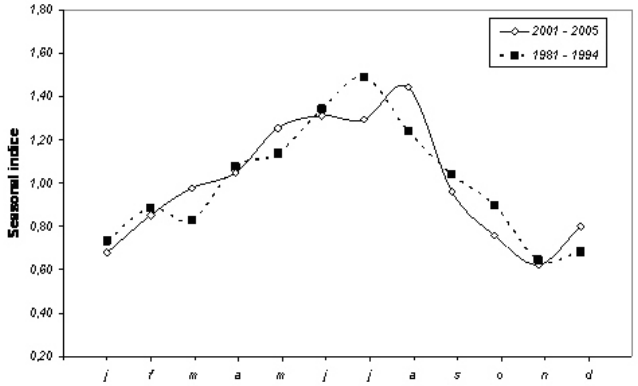

Fig. 5. Seasonal indices (ratio of monthly mean and overall average of activity concentration) of activity concentration of ^{7}Be in low-level atmosphere in Bratislava for two time periods, 1981–1994 and 2001–2004.

tween the air reservoirs with consequent transport of cosmogenic radionuclides to low-level atmosphere. The intrusions of stratospheric air into the troposphere are processes occurring during the upward movement of tropopause. On the contrary in cold months these exchange processes are reduced and as a result the supply of ^{7}Be produced in higher layers of atmosphere declines.

In measured data appeared a new feature in typical seasonal variation pattern. It is the local minimum in summer, in June or July, evident from the comparison of seasonal variations of activity concentrations of ^{7}Be in Bratislava in periods 1981–1994 and 2001–2004 (Fig. 4). The older data does not indicate such a feature observable in present measurements. Similar trend is observable in data from Prague [10] and from other locations in Slovakia. It may indicate the modification of character of weather summer season in middle Europe region. On the territory of Slovak Republic are few institutions which are monitoring the radioactivity of aerosol content of atmosphere. Besides the laboratory at the Department of Nuclear Physics and Biophysics, there are the Laboratories of Radiation Control of Vicinity of Nuclear Power Plants (NPP) Jaslovske Bohunice and Mochovce, and Slovak Hydro-Meteorological Institute (SHMI). In the surroundings of NPP J. Bohunice are 8 stations, near NPP Mochovce are 13 monitoring stations and under authority of SHMI come 3 stations. Apart from other radionuclides is in these monitoring stations also cosmogenic ^{7}Be determined. It has to be taken into consideration that main reason of measurements is continual monitoring of neighbourhood of power plants. However, for purposes to show trends of seasonal indices are this data fully suitable.

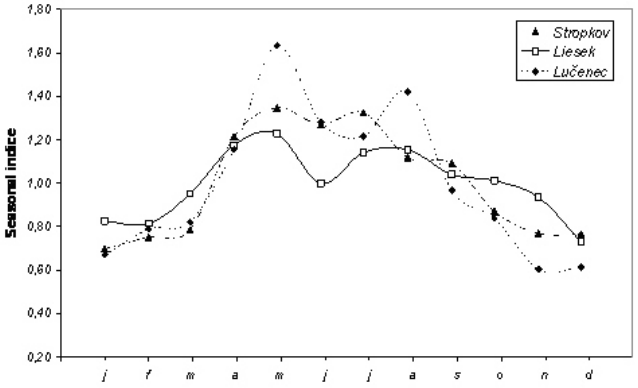

Fig. 6. Seasonal indices of activity concentration of ^{7}Be for three stations under authority of SHMI for period 1998–2003 in Stropkov, 1996–2003 (without year 1998) in Lučenec and 1996–2003 (without years 1999–2001) in Liesek.

Figs. 6–8 present seasonal indices of ^{7}Be activity concentrations in three stations under the authority of SHMI, five selected stations in the neighbourhood of NPP J. Bohunice and five selected locations near NPP Mochovce. The local minimum is clearly observable in the figures in summer. We suppose this effect is connected with changes of global climate and associated changes of atmospheric circulation. The air masses forming tropic and subtropic area are shifted to the middle latitude more intensively. Concentrations of cosmogenic radionuclides in the middle latitude locations descend owing to lower concentrations in these

air masses compared to air originating from higher latitudes. Full explanation needs a long-term monitoring of activity concentration of ^{7}Be and further study of this problem.

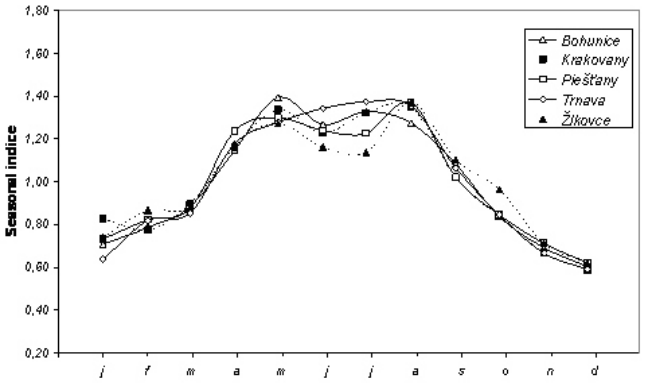

Fig. 7. Seasonal indices of activity concentration of ^{7}Be for period 1995–2002 in five stations in the vicinity of NPP Jaslovské Bohunice.

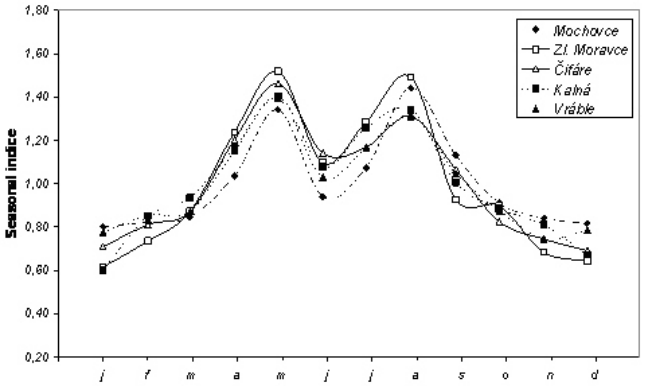

Fig. 8. Seasonal indices of activity concentration of ^{7}Be in for period 1998–2002 in five stations in the vicinity of NPP Mochovce.

Table 1. Activity concentrations of ^{7}Be in low-level atmosphere in 14 sampling locations in Slovakia.

	Location	**Period**	**Concentration** [mBq.m ^{3}]	**Average** [mBq.m ^{3}]
	Bratislava	1981–1994	0.70–8.69	3.15 ± 0.10
		2001–2005	0.59–5.28	2.43 ± 0.10
NPP J. Bohunice	J. Bohunice	1995–2002	0.67–5.18	2.45
	Krakovany		0.43–5.48	2.63
	Pieš any		0.68–5.18	2.13
	Trnava		0.73–10.37	3.12
	Žlkovce		0.33–8.41	2.97
NPP Mochovce	Zl. Moravce	1998–2002	0.28–6.65	3.04
	Čifáre		0.27–5.77	2.70
	Kalná		0.30–6.61	3.04
	Mochovce		0.52–6.60	2.81
	Vráble		0.58–5.66	2.75
SHMI	Stropkov	1998–2003	0.23–4.10	1.68
	Lučenec	1996–1997, 1999–2003	0.52–3.70	1.72
	Liesek	1996–1998, 2002–2003	0.15–3.84	1.46

Table 2. Activity concentrations of ^{7}Be in low-level atmosphere in six different locations.

Location	**Period**	**Concentration** [mBq.m ^{3}]	**Mean** [mBq.m ^{3}]
Neuherberg – Germany [15]	1994–1996	1.40–6.70	3.33
Granada – Spain [7]	1993–2001	1.50–7.60	4.45 ± 1.35
Monaco [16]	1997–2000	1.50–7.24	4.21
El-Minia – Egypt [11]	1998–1999	1.10–3.00	2.0 ± 0.09
Prague [10]	1986–2002	1.01–7.07	2.92

Table 2 presents activity concentrations of ^{7}Be in six different locations. The concentration of ^{7}Be measured in Bratislava compared to other locations seems to be low, but it is a result of anticorrelation between the production of cosmogenic radionuclides and the sun activity. Monitored period 2001–2004 does not cover the entire 11-year sun cycle, but only the period with increased sun activity.

4. Conclusion

The concentrations of radionuclide ^{7}Be for the monitoring period 2001–2005 ranged from 0.59 to 5.28 mBq.m ^{3} with the mean value (2.43 ± 0.10) mBq.m ^{3} and show seasonal variations. Activity of aerosol component of low level atmosphere in Bratislava shows typical values of air activity concentrations of ^{7}Be for European area. The newly observed local

summer minimum can be explained as the result of climatic changes when the intensity of horizontal advection increases. This problem needs more analyses and further studies.

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References

[1] D. Lal, B. Peters: Cosmic Ray Produced Radioactivity in the Earth. Hand. Physics **46** (1967) 551 612.
[2] C. Papastifanou, A. Ioannidou: Aerodynamic Size Association of ^{7}Be in Ambient Aerosols. Journal of Environmental Radioactivity **26** (1995) 273 282.
[3] G. M. Raisbeck, F. Yiou: Cosmogenic ^{10}Be/^{7}Be as a Probe of Atmospheric Transport Processes. Geophysical Research Letters **8** (1981) 1015 1018.
[4] J. Roed, R. J. Cannel: Relationship between Indoor and Outdoor Aerosol Concentration Following the Chernobyl Accident. Radiation Protection Dosimentry **21** (1987)107 110.
[5] H. W. Feely, R. J. Larsen, C. G. Sanderson: Factors that Cause Seasonal Variations in Beryllium-7 Concentrations in Surface Air. Journal of Environmental Radioactivity **9** (1989) 223 249.
[6] D. Al-Azmi, A. M. Sayed, H. A. Yatim: Variations in Be-7 Concentrations in Atmosphere of Kuwait during the Period 1994 to 1998. Applied Radiation and Isotopes **55** (2001) 413 417.
[7] M. Azahra, A. Camacho-García, C. González-Gómez, J. J. López-Peñalver, T. El Bardouni: Seasonal ^{7}Be Concentrations in Near-Surface Air of Granada (Spain) in the Period 1993–2001. Applied Radiation and Isotopes **59** (2003)159 164.
[8] F. Cannizzaro, G. Greco, M. Raneli, M. C. Spitale, E. Tomarchio: Concentration measurements of Be-7 at Ground Level Air at Palermo, Italy - Comparison with Solar Activity over a Period of 21 years. Journal of Environmental Radioactivity **72** (2004) 259 271.
[9] D. Todorovic, D. Popovic, G. Djuric, M. Radenkovic: Be-7 to Pb-210 Concentration Ratio in Ground Level Air in Belgrade Area. Journal of Environmental Radioactivity **79** (2005) 297 307.
[10] P. Kuča, L. Novák, P. Rulík, J. Tecl: Radiation Monitoring Network of the Czech Republic. Proceedings of the IRPA Regional Congress on Radiation Protection in Central Europe, Bratislava, Slovakia, September **22 26** (2003).
[11] A. El-Hussein, A. Mohamemed, M. Abd EL-Hady, A. A. Ahmed, A. E. Ali, A. Barakat: Diurnal and Seasonal Variation of Short-lived Radon Progeny Concentration and Atmospheric Temporal Variations of ^{210}Pb and ^{7}Be in Egypt. Atmospheric Environment **35** (2001) 4305 4313.
[12] L. Ďurana, M. Chudý, J. Masarik: Investigation of Be-7 in the Bratislava Atmosphere. Journal of Radioanalytical and Nuclear Chemistry **207** (1996) 345 356.
[13] I. Sýkora, M. Chudý, L. Ďurana, K. Holý, J. Merešová: Aerosols Radioactivity in the Bratislava Atmosphere. Conf. Proc. of the 24th Days of Radiation Protection, Jasná pod Chopkom, Slovakia, (2001) 145 147.
[14] I. Sýkora, J. Merešová, M. Chudý, K. Holý: The Study of Aerosol Component of Atmosphere in Bratislava. Proceedings of the IRPA Regional Congress on Radiation Protection in Central Europe, Bratislava, Slovakia, September 22 26, 2003.
[15] R. Winkler, F. Dietl, G. Frank, J. Tschiersch: Temporal Variation of ^{7}Be and ^{210}Pb Size Distributions in Ambient Aerosol. Atmospheric Environment **32** (1998) 983 991.
[16] S.-H. Lee, M. K. Pham, P. Povinec: Radionuclide Variations in the Air over Monaco. Journal of Radioanalyt. and Nucl. Chemistry **254** (2002) 445 453.