GROUND LEVEL AIR BERYLLIUM-7 AND OZONE IN BELGRADE

by

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Three sets of data covering the 2004-2007 period are examined: two beryllium-7 series and ozone measured in ground level air. The measuring sites are at three different locations in Belgrade, Serbia. The temporal evolution of beryllium-7 and ozone is presented, as well as their mutual correlations. Beryllium-7 data for Belgrade agree well with the results for other locations in the region. The correlation between two beryllium-7 data sets is 0.57. The results for ozone indicate that Belgrade is not a common continental site, as the maximum in ozone distribution is reached in springtime. The overall correlation between beryllium-7 and ozone is good, but varies over different seasons. A large correlation (0.67) is noted between beryllium-7 measured at the site in Vinča, Serbia, and the monthly maximum ozone in autumn. An analysis which assumes the transport of air masses from the stratosphere, along which the only process changing the air mass composition is radioactive decay of beryllium-7, does not conclusively confirm the high correlation between beryllium-7 and ozone in autumn.

Key words: ground level air, beryllium-7, ozone, stratosphere-troposphere exchange

INTRODUCTION

Beryllium-7 (^{7}Be) and ozone (O_3) are atmospheric constituents which have distinct concentrations and behaviour in the stratosphere and the troposphere.

^{7}Be is a radioactive element (half-life 53.3 days) produced by cosmic rays in spallation processes, in the lower stratosphere (~70%) and the upper troposphere (~30%) [1]. Following production, ^{7}Be is promptly attached to aerosols with a diameter of 0.3-0.6 m whose residence time in the atmosphere is around 20 days [2]. In the troposphere, apart from its decay, ^{7}Be is removed by wet deposition (the major mechanism for removal) and dry deposition [1]. In ground level air, the residence time of ^{7}Be is around 10 days [3].

Ground level ^{7}Be has been measured in Belgrade, Serbia, since 1996, as a part of the air radioactivity monitoring programme in the Vinča Institute of Nuclear Sciences. Todorovic *et al.* [4, 5] reported that the measured values of ^{7}Be in Belgrade air are in good agreement with the data for other regions, and showed that the ^{7}Be activity reaches its maximum in summer.

Most of O_3 in the atmosphere is produced in the stratosphere [6]. In the troposphere, O_3 is formed in photochemical reactions and its distribution is strongly dependant on the availability of source gases, as well as meteorological conditions [7, 8]. The average amount of ozone in the troposphere is approximately 10% of the total ozone [9], but it can be as low as 5% in unpolluted areas [10]. Ground level ozone is of special interest because of its adverse effects on ecosystems and the environment in general [9].

According to numerous studies which have reported the O_3 annual cycle, Monks [11] classified the measuring sites into two groups: the ones that have a broad maximum in summer and the ones that have a

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spring maximum. Continental sites influenced by pollution, whereby photochemical production of ozone is enabled, comprise the first group [11]. Likewise, Di Carlo *et al*. [12] reported a July maximum in ground level O_3 measured in central Italy. Although there are no data for the Balkans in Monks' [11] report, it is expected that Belgrade should fall into this group. The spring maximum (May) was noted at coastal sites, such as along the North and the Baltic Sea [11].

Coincidental measurements of ^{7}Be and O_3, both of which can be used as stratospheric tracers [13-16], have proved to be very informative of the atmospheric processes. The tropospheric budget of ^{7}Be and O_3 is influenced by the stratosphere-troposphere exchange (STE), whereby stratospheric air masses enter the troposphere, thus changing the composition of the troposphere [17]. Since ^{7}Be and O_3 concentrations are much higher in the stratosphere, an intrusion of stratospheric air can be detected through elevated concentrations of these constituents in the troposphere [18].

In the uppermost troposphere, an STE event can be readily discerned before the transported air mass loses its stratospheric characteristics. This phenomenon has been documented in a number of studies [19-21]. On the other hand, an analysis of an aged stratospheric intrusion is more complicated, since various mechanisms in the troposphere diminish the stratospheric signature [19]. Fewer STE events were detected in the middle troposphere [1, 22], because by the time the lump of stratospheric air reaches those altitudes, the signatures are usually gone. Furthermore, there are examples of deep stratospheric intrusions reaching the surface [23, 24].

Despite the difficulties in identifying air of stratospheric origin at the surface, some studies used the correlation between ^{7}Be and O_3 to quantify the contribution of stratospheric O_3 to ground level O_3 [13] or to examine whether coincidental elevated levels of ^{7}Be and O_3 were caused by an intrusion of stratospheric air [25].

In this paper, concentrations of ^{7}Be and O_3 measured at different locations in Belgrade are presented. The results for ^{7}Be and O_3 are first discussed separately, in order to examine whether Belgrade is an uncommon site. The correlation between ^{7}Be and O_3 is also given and the results discussed in detail.

MATERIALS AND METHODS

Aerosol samples for ^{7}Be measurements were collected using constant flow rate samplers (average air flow of 25 m^3/h) and FILTRAK/WHATMAN 41/DDR filter papers (15 cm in diameter, with a relative efficiency for deposited dust of 80%), at two locations: at the Vinča Institute of Nuclear Sciences (hereinafter, Vinča), in a Belgrade suburb, and in Karadjordjev park (hereinafter, KP), in the centre of the city. Monthly composite samples (average volume of 15 10^3 m^3) were formed after ashing at temperatures below 400 °C. The monthly samples were then placed in small metallic containers (inner diameter of 25 mm, outer diameter of 27 mm and height of 4 mm) and analysed [4].

The activity of ^{7}Be was determined on two Canberra HPGe detectors (with relative efficiencies of 20% and 23%) by standard gamma spectrometry. Energy calibration was preformed with a set of standard point sources (Coffret d'etalon gamma ECGS-2, Sacle, France) containing ^{133}Ba, ^{57,60}Co, and ^{137}Cs. Geometric efficiency was determined with the IAEA-83 (AIR4) simulated air filter (a filter paper spiked with a solution of ^{60}Co, ^{90}Sr, ^{133}Ba, ^{137}Cs, and ^{210}Pb; ^{210}Pb activity 155 Bq, with an uncertainty of 5%). Mathematical approximations were used to estimate the efficiency for the geometry of the composite sample. Counting time intervals were between 150 000 s and 250 000 s. The background spectrum was regularly recorded prior to or after sample counting. The total standard error of the method (including the relative uncertainties in geometric efficiency estimation, photopeak counts estimation, sample volume determination, *etc*.) was estimated to be below 25%. The lower detection limit for ^{7}Be was 1 10^{-5} Bq/m^3 [4].

Ozone measurements were performed by the APOA-360 HORIBA Ambient O_3 Monitor (the method based on UV 254 nm absorption: DIN ISO 13964 "Ambient air quality – Determination of ozone in ambient air – ultraviolet photometric method"). The lower detectable limit of the method is 0.4 parts per billion (2σ). The monitor is located at a site in New Belgrade, approximately 10 km away from the city centre (KP). Hourly average ozone data covering the 2004-2007 period are available.

RESULTS AND DISCUSSION

Monthly mean values of ^{7}Be measured at two sites (KP and Vinča) and O_3 (measured in New Belgrade) over the 2004-2007 period are given in fig. 1. The two ^{7}Be data sets show a good agreement, although in 2004 and at the beginning of 2005, ^{7}Be in Vinča was consistently higher than ^{7}Be in KP. The linear Pearson correlation coefficient for the two ^{7}Be data sets is 0.57.

^{7}Be and O_3 both show a very similar annual cycle, which can be seen more clearly in figs. 2 and 3. The maximum in ^{7}Be concentration in Vinča is reached in May and June, while the maximum at KP is broader (includes the month of April). Both data sets show the minimum in late autumn and winter (fig. 2). Ozone monthly distribution shows the maximum in May, with high values throughout spring and summer, a slow decrease in autumn, followed by a relatively sharp increase in winter (fig. 3).

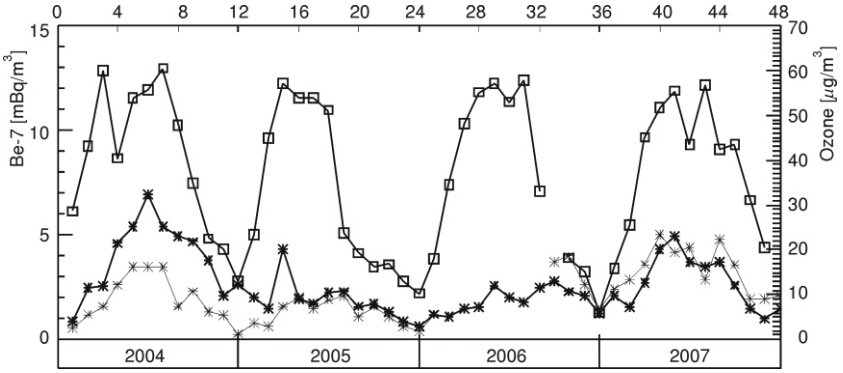

Figure 1. Monthly mean values of ^{7}Be in Vinča (thick asterisks) and KP (thin asterisks), and O_3 (squares) over 2004-2007; data points for February through to August 2006 are missing in the KP ^{7}Be data set, as well as September 2006 in the O_3 data set

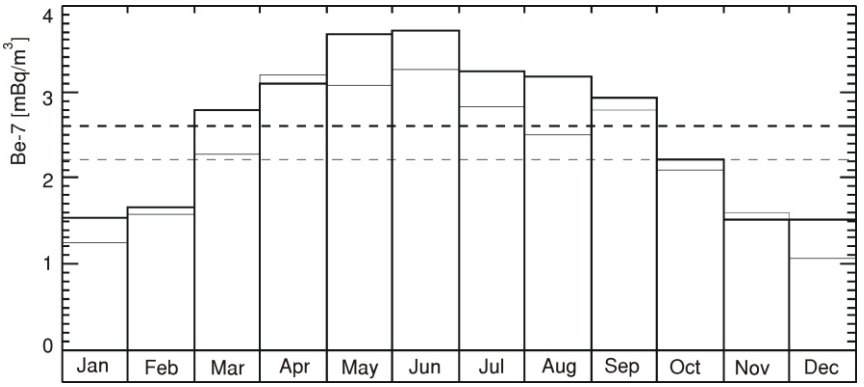

Figure 2. ^{7}Be monthly mean values over the 2004-2007 period in Vinča (thick lines) and KP (thin lines); the thick dashed and thin dashed lines represent the overall mean (the average value over the examined period) for ^{7}Be values for Vinča and KP, respectively

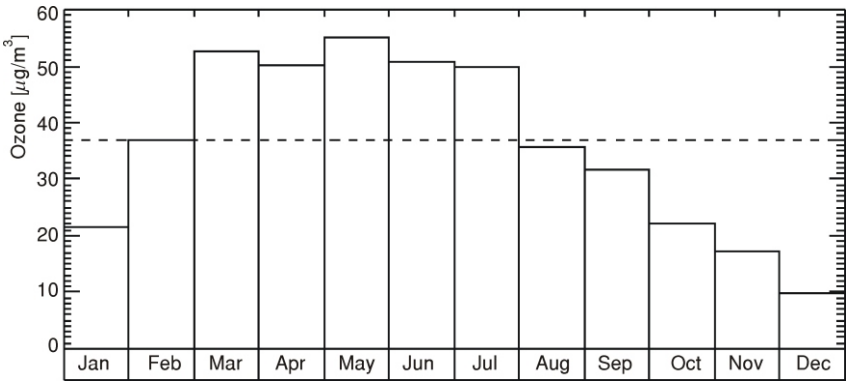

Figure 3. O_3 monthly mean values over the 2004-2007 period; the dashed line represents the overall mean (the average value over the examined period) O_3 value

These results agree well with other Belgrade studies [4, 5] and with results obtained at other locations in Europe. For example, Ďurana *et al*. [26] presented the 1977-1994 results for the Bratislava atmosphere, showing that the ^{7}Be activity rarely exceeded 6 mBq/m^3, and that it reached the maximum in June. Likewise, over 1993-1997 Azahra *et al*. [27] measured ^{7}Be in Granada, and demonstrated that the highest values were reached in summer. Similar results for ground level ^{7}Be were shown by Irlweck *et al*. [25] and others [2, 16, 28-30].

The monthly mean O_3 values in the examined period were below 70 μg/m^3 (fig. 1). However, the monthly maximum for O_3 (not shown in fig. 1) exceeded the 150 μg/m^3 yearly summer threshold. Therefore, a more detailed analysis should be performed in order to reach sound conclusions about ground level O_3 behaviour in Belgrade.

The O_3 annual cycle is obvious in figs. 1 and 3, with minima in winter and maxima in May. In this respect, Belgrade does not fall into the category of polluted continental sites [1, 11]. The springtime maximum is considered a consequence of transport and photochemistry interplay [1, 11]. The four examined years might, however, be insufficient to draw a definite conclusion. An extension of the O_3 data set should resolve whether the May maximum, higher by approximately 5% than the means in other spring and summer months (fig. 3), is only a part of a very broad spring-summer maximum.

To further analyse the behaviour of ^{7}Be and O_3, seasonal indices were calculated [5]. A monthly seasonal index represents the ratio of the monthly mean value and the overall average value for the examined period. A quarterly seasonal index is the

Table 1. Seasonal indices for ^{7}Be in Vinča and at KP and for O3, 2004-2007

Month	12	1	2	3	4	5	6	7	8	9	10	11
^{7}Be (Vinča)	0.58	0.60	0.64	1.08	1.20	1.42	1.43	1.25	1.23	1.13	0.86	0.58
^{7}Be (KP)	0.46	0.54	0.69	0.99	1.40	1.34	1.43	1.24	1.09	1.22	0.91	0.70
O_3	0.27	0.60	1.03	1.45	1.39	1.53	1.41	1.38	0.99	0.87	0.61	0.48
Quarter	Winter			Spring			Summer			Autumn		
^{7}Be (Vinča)	0.61			1.23			1.30			0.86		
^{7}Be (KP)	0.56			1.24			1.25			0.94		
O_3	0.63			1.46			1.26			0.65		

mean of the three monthly indices for the corresponding season, *i. e*., December, January, and February for winter, March, April, and May for spring, June, July, and August for summer, and September, October, and November for autumn. The seasonal indices are given in tab. 1.

As can be seen from tab. 1, both ^{7}Be and O_3 concentrations are higher than the overall mean, but there is a lag in ^{7}Be concentration compared to O_3 concentration: O_3 concentration becomes higher than the overall mean in February, and drops below the mean in September, while ^{7}Be concentration shows this same behaviour in March and October.

To quantify the correlation between ^{7}Be and O_3, linear Pearson correlation coefficients were calculated for the whole set of experimental data, as well as for different seasons. The results are given in tab. 2.

The overall correlation (columns denoted by "All" in tab. 2) between ^{7}Be and O_3, both mean and maximum, is good. The correlation for the mean O_3 is poor in winter and spring, and also in summer for the maximum O_3. A strikingly large correlation exists between ^{7}Be at the Vinča site and the maximum O_3 in autumn.

Correlation coefficients between ^{7}Be and O_3 have been previously reported in a number of studies. For example, Allen *et al*. [1] obtained the ^{7}Be/O_3 correlation coefficients in the range of 0.5-0.7 for the lower troposphere (only free troposphere was examined in the study), and concluded that in the tropospheric layer ^{7}Be can be used as a tracer, even if not a very good one. Our results, however, show that in spring, in the boundary layer, ^{7}Be is not significantly correlated with O_3.

The large correlation coefficient in autumn (tab. 2) could indicate that ^{7}Be and the maximum O_3 are governed by the same processes. One possible explanation is the existence of the downward transport from higher altitudes. However, the monthly mean values of ^{7}Be and O_3 in autumn (fig. 1) do not show an obvious increase in either of the concentrations (the same stands for the maximum O_3), which would support the assumption of stratospheric air reaching lowermost parts of the troposphere. On the other hand, the monthly mean values are probably too coarse to directly show an intrusion of air from the stratosphere. Daily and hourly values are more appropriate and have been widely used [19, 25]. Our study, however, is limited by the fact that only the monthly mean ^{7}Be data were available.

Nevertheless, assuming that during the autumn months, some of the air masses sampled in Belgrade originated in the stratosphere, a relatively simple analysis was performed using the monthly mean values. The analysis consisted of three basic steps. In the first step, it was assumed that a stratospheric air mass, characterized by a constant value of the ^{7}Be/O_3 ratio, entered the troposphere. In the second step, it was assumed that the descent of the stratospheric air mass took one month, during which the stratospheric ^{7}Be/O_3 ratio was changed solely due to the ^{7}Be decay. Finally, in the last step, it was assumed that at the surface, where the air mass was sampled, a tropospheric value of the ^{7}Be/O_3 ratio could be used to identify the air mass of stratospheric origin. Therefore, one of the aims of the analysis was to obtain the tropospheric ^{7}Be/O_3 ratio of stratospheric air masses.

To calculate the tropospheric ^{7}Be/O_3 ratio, the results of two studies were used. Dutkiewicz and Husain [13] showed that over the April-June period, the average stratospheric ^{7}Be/O_3 ratio is 0.4 mBq/m^3 per parts per billion by volume (0.2 mBq/μg). On the other hand, Stohl *et al*. [19] examined a prominent STE event over three Alpine peaks in October 1996,

Table 2. Linear Pearson correlation coefficients of ^{7}Be and O_3; the coefficients with the absolute value belonging to [0.3, 0.5) and [0.5, 1.0], indicating a medium and a large correlation, are given in italic bold, and bold, respectively

7Be	Mean O_3					Maximum O_3				
	All	Winter	Spring	Summer	Autumn	All	Winter	Spring	Summer	Autumn
Vinča	**0.52**	0.13	–0.10	***0.47***	**0.51**	***0.49***	0.26	–0.13	0.26	**0.67**
KP	***0.47***	–0.05	–0.28	***0.42***	***0.46***	***0.36***	–0.16	***–0.31***	0.17	***0.33***

and, according to their measured concentrations of ^{7}Be and O_3, the ^{7}Be/O_3 ratio varied between approximately 0.1 mBq/μg and 0.155 mBq/μg. These three values were used in our analysis as the stratospheric ^{7}Be/O_3 ratios.

Once in the troposphere, the composition of stratospheric air mass changes as a consequence of a number of processes which independently influence the amount of ^{7}Be and O_3. ^{7}Be decays (a primary mechanism for loss) and is removed from the atmosphere by wet scavenging (according to Allen *et al*. [1], dry deposition plays a minor role). Ozone, on the other hand, undergoes chemical reactions which can either lead to O_3 production or destruction, depending on the irradiation and the availability of ozone precursors [11]. The photochemical life time of O_3 in the troposphere is long during months of low irradiation [31], *i. e*. in autumn and winter. In our analysis, therefore, it was assumed that the initial stratospheric value of ^{7}Be/O_3 ratio was decreased only through ^{7}Be decay.

Considering that the descent of stratospheric air masses takes about a month [14], and assuming that the wet scavenging of ^{7}Be and mixing of air masses can be neglected over that period, the tropospheric ^{7}Be/O_3 ratios were calculated. The ratio of 0.2 mBq/μg from Dutkiewicz and Husain [13] gave 0.134 mBq/μg as a tropospheric ^{7}Be/O_3 ratio, and the limits from Stohl *et al*. [19] of 0.1 mBq/μg and 0.155 mBq/μg gave 0.067 mBq/μg and 0.104 mBq/μg, respectively. Using these ratios, air of stratospheric origin could be identified even if there were no obvious peaks in ^{7}Be and O_3 concentrations.

The ^{7}Be/O_3 ratios over the 2004-2007 period, calculated from our measurements, are given in fig. 4. The ratios were calculated for the monthly mean O_3 and for the monthly maximum O_3. Comparing the ratio of ^{7}Be and mean O_3 to the ratio value based on Dutkiewicz and Husain [13], it could be concluded that air sampled during September, October, and December 2004, and during November and December 2006, carried the stratospheric characteristics. The upper ratio limit based on Stohl *et al*. [19] also included November 2004 and September 2005 as candidate months (as well as April and June 2004). The lower ratio limit based on Stohl *et al*. [19], on the other hand, indicated many occurrences of stratospheric air sampling.

The monthly mean O_3, however, is probably not a good representative of the assumed unchanged stratospheric O_3, as it is too liable to decrease caused by mixing of tropospheric air. Therefore, the ratio of ^{7}Be and the monthly maximum O_3 was also examined. This ratio was below the lower ratio limit based on Stohl *et al.* [19] throughout the examined period. Finally, the monthly mean of daily maxima of O_3 for the autumn months was taken as a possible representative of stratospheric O_3, which gave October 2004 as a candidate month for sampling air of stratospheric origin.

The above analysis does not conclusively explain the large correlation between ^{7}Be and the maximum O_3. In fact, only the examination of the ^{7}Be and the monthly mean O_3 ratio confirmed the premise of an aged stratospheric air. There is, therefore, a need to refine the assumptions in many respects. For example, the trajectories of air sampled in Belgrade during autumn could be closely examined, along with the amount of precipitation that the sampled air underwent along the trajectories. Moreover, the stratospheric ^{7}Be/O_3 ratio is of crucial importance in the proposed method. Hourly and daily measurements of ^{7}Be and O_3, conducted at one measuring site, would also aid to verify the origin of air masses in Belgrade.

Nevertheless, it is worth mentioning that the basic assumption of the analysis is valid. Stohl *et al*. [19] showed that the stratospheric intrusions above the Alpine peaks are most frequent in October, and that their direct influence is stronger in the regions east of the Alps. Therefore, it is reasonable to expect that at least some of the stratospheric air masses reach Belgrade in autumn. Furthermore, even though it was not shown in the study of Stohl *et a*l. [19], they argued that stratospheric O_3 is not a negligible contributor to the boundary layer O_3. In other words, Belgrade may prove to be a very good site to confirm the stratospheric influence on ground level O_3.

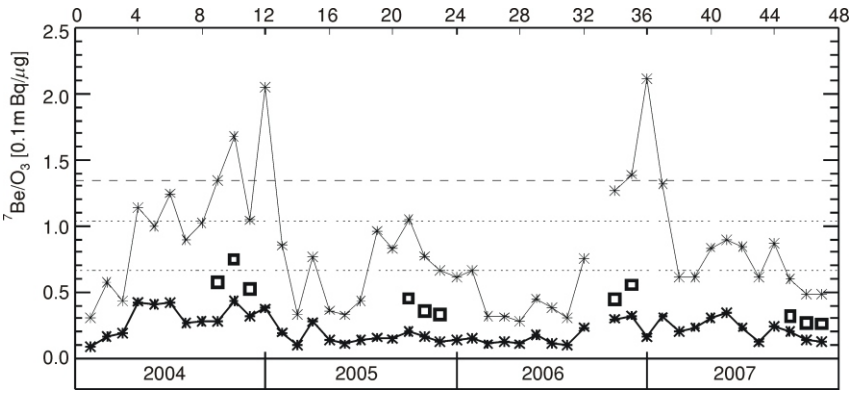

Figure 4. Ratio of monthly mean ^{7}Be measured in Vinča and monthly mean and monthly maximum O_3 (thin and thick asterisks, respectively); the ratio of ^{7}Be and monthly mean maximum (monthly mean of daily maximum values) for the autumn months is given in squares; the dashed line represents 0.134 mBq/μg, calculated according to Dutkiewicz and Husain [13]; the two dotted lines represent the ratio limits of 0.067 mBq/μg and 0.104 mBq/μg, calculated according to Stohl *et al.* [19]

CONCLUSIONS

Time series of ^{7}Be and O_3, measured at three different locations in Belgrade over the 2004-2007 period, were presented. The ^{7}Be results show that Belgrade is similar to other urban sites, while the May maximum in the O_3 data indicates that Belgrade does not fall into the category of urban continental sites characterized by a broad summer maximum. Additional O_3 data are needed to confirm this conclusion.

The correlation between the data sets was also examined. The correlation coefficient between the two ^{7}Be sets is 0.57. On the other hand, the correlation between ^{7}Be and mean O_3 is not as large and varies with season. A large correlation coefficient of 0.67 was calculated for ^{7}Be and the monthly maximum O_3 in autumn, which implies that their concentrations are driven by the same processes.

To explain the high ^{7}Be to ozone correlation in autumn, a relatively simple analysis was performed. It was assumed that an aged stratospheric air was sampled in Belgrade during autumn, and that the only mechanism changing the air mass composition along its descent was the radioactive decay of ^{7}Be. The analysis confirmed the initial premise, but not conclusively, and a more detailed study should be performed.

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БЕРИЛИЈУМ-7 И ОЗОН У ПРИЗЕМНОМ СЛОЈУ АТМОСФЕРЕ У БЕОГРАДУ

У периоду од 2004. до 2007. године на три локације у Београду мерени су берилијум-7 и озон у приземном слоју атмосфере. Берилијум-7 мерен је у Винчи и Карађорђевом парку, а озон у Новом Београду. Анализиране су сезонске промене концентрације берилијума-7 и озона у ваздуху и њихов међусобни однос. Добијене вредности за берилијум-7 у Београду не одступају од резултата добијених на другим локацијама у региону. Корелација између две групе података за берилијум-7 (локација Винча и локација Карађорђев парк) је 0,57. Добијени резултати за озон указују да Београд није уобичајена континентална локација, јер се максимум дистрибуције озона јавља у пролеће. Корелација између берилијума-7 и озона током читавог мерног периода је добра, али постоје разлике по годишњим добима. Висока корелација (0,67) добијена је током јесени за берилијум-7 (локација Винча) и максималне месечне концентрације озона. Висока корелација између добијених вредности за озон и берилијум-7 у приземном слоју атмосфере током јесењих месеци не може се у потпуности објаснити само транспортом ваздушних маса из стратосфере, при чему је једини процес који мења састав ваздушних маса радиоактивни распад берилијума-7.

Кључне речи: приземни ваздух, берилијум-7, озон, размена стратосфере и тропосфере