

# A MULTI-YEAR STUDY OF RADIOACTIVITY IN SURFACE AIR AND ITS RELATION TO CLIMATE VARIABLES IN BELGRADE, SERBIA

by

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Scientific paper

DOI: 10.2298/NTRP1304381A

Activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  were monitored in surface air in Belgrade, Serbia, from 2004 to 2012. The measurements were taken from two locations, in an open field of a city suburb and in the central city area. The activities were determined on HPGe detectors by standard gamma spectrometry. The  $^7\text{Be}$  activity shows a pronounced seasonal pattern, with the maximum in spring-summer and minimum in winter, while the  $^{210}\text{Pb}$  activity exhibits two maxima, in autumn and late winter. The mean monthly concentrations measured at both sites are below  $9 \text{ mBq/m}^3$  and  $1.3 \text{ mBq/m}^3$  for  $^7\text{Be}$  and  $^{210}\text{Pb}$ , respectively. The obtained correlation of the  $^7\text{Be}$  activity with the number of sun-spots is not statistically significant. Relations of the radionuclides' activities with climate variables (precipitation, temperature, relative humidity, cloud cover, sunshine hours, and atmospheric pressure) are also investigated, but the only significant correlations are found for the  $^7\text{Be}$  activity with temperature and sunshine hours, and the  $^{210}\text{Pb}$  activity with atmospheric pressure. The maximum  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities corresponding to binned total monthly precipitation data imply different modes of the radionuclide scavenging from the atmosphere. During dry periods, accumulation of the radionuclides in the atmosphere leads to their increased activities, but no correlation was found between the activities and the number of consecutive dry days.

*Key words: radioactivity, beryllium-7, lead-210, air, sun-spot number, climate variable*

## INTRODUCTION

Air radioactivity monitoring is an important part of environmental radioactivity monitoring, as it provides data on the content of the radionuclides, their origin and production rate in the atmosphere. The monitoring also plays a vital role in understanding the atmospheric dynamics processes, such as the stratosphere-to-troposphere exchange and circulation within the troposphere. The most extensively monitored radionuclides in the air are the cosmogenic  $^7\text{Be}$ , long-lived radon progeny  $^{210}\text{Pb}$ , and anthropogenic  $^{137}\text{Cs}$ .

Beryllium-7 (half-life 53.28 days) is produced in the upper troposphere and lower stratosphere, where cosmic rays interact with light elements [1]. The average concentration of  $^7\text{Be}$  in the troposphere is  $12.5 \text{ mBq/m}^3$  [2]. The  $^7\text{Be}$  seasonal pattern in the

troposphere, showing the maximum in the spring/summer season, is correlated with the intrusion of the stratospheric air masses across the tropopause [3-6]. Monthly  $^7\text{Be}$  activities in surface air are inversely correlated with solar activity [7, 8]. Different studies investigated the correlation of the  $^7\text{Be}$  activity with precipitation and their results showed no correlation or a negative correlation [7-12]. However, a good positive correlation between  $^7\text{Be}$  monthly deposition and the amount of precipitation was found by [13, 14]. As a good tracer of air mass origin,  $^7\text{Be}$  is also a good indicator of injection of ozone and aerosols from the upper into the lower atmosphere [15, 16].

Lead-210 (half-life 22.3 years) is a member of  $^{222}\text{Rn}$  decay series (a decay product of  $^{238}\text{U}$ ). Small amounts of  $^{210}\text{Pb}$  found in surface air are due to: (1) decay of  $^{222}\text{Rn}$  residing in the air, (2) ground resuspension, and (3) anthropogenic activities, mainly coal combustion. Concentrations of  $^{210}\text{Pb}$  in the air

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generally exhibit maxima during autumn, caused by an increased emanation of radon [17-21]. On the global scale, higher concentrations of  $^{210}\text{Pb}$  are found in mid-latitudes, while on the local level, emanation of radon from the soil, and therefore concentrations of  $^{210}\text{Pb}$  in the air, are strongly affected by atmospheric pressure, temperature, vegetation and snow cover [22-24].

Due to their different origin, the concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  vary with height in the atmosphere. Air masses coming from the upper troposphere and lower stratosphere contain higher concentrations of  $^7\text{Be}$  than the surface air masses. On the other hand, surface air masses are richer in  $^{210}\text{Pb}$  than air masses originating from the higher altitudes. Furthermore, the  $^{210}\text{Pb}$  concentration is higher in continental air masses than in air masses originating over a body of water [25]. Both of the radionuclides have a long residence time in the atmosphere [26-28]. The activity ratio  $^7\text{Be}/^{210}\text{Pb}$  strongly depends on the altitude at which the air is transported, thus providing information on the rate and the velocity of the air convection processes [29]. Since both radionuclides have higher concentrations during warm seasons, when convection of the air moves  $^{210}\text{Pb}$  upwards and  $^7\text{Be}$  downwards, the  $^7\text{Be}/^{210}\text{Pb}$  ratio exhibits summer maxima and winter minima [8, 26, 30]. The radionuclides have similar particle size distributions and therefore, their mean radioactive loading index, which describes their relative abundance, is rather stable on the global scale [31]. Washout is the major removal process from the atmosphere for both  $^7\text{Be}$  and  $^{210}\text{Pb}$  [14].

The activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in Serbia have been studied previously [3, 16, 21, 32-35]. The results of the studies showed that the average monthly concentrations of  $^7\text{Be}$  in surface air were between  $2.0 \text{ mBq/m}^3$  and  $10.0 \text{ mBq/m}^3$ , with a pronounced maximum in summer or early autumn and a minimum in winter, and the  $^{210}\text{Pb}$  concentrations in the range of  $0.25\text{-}3.40 \text{ mBq/m}^3$ , with the maximum in autumn.

In this paper, the results of a comparative study of the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities at two sites in the city of Belgrade are given. During 2004-2012, the activities were measured in an open field location, within the Vinča Institute of Nuclear Sciences (hereinafter, Institute), and in a central urban area location (hereinafter, City). The aim of the study was to examine the concentrations of the radionuclides and their correlation across the sites. In addition, the influence of solar activity and local climate, with a special emphasis given to precipitation, was investigated.

## MATERIALS AND METHODS

The samples of air were collected in the city of Belgrade ( $44.80^\circ \text{ N}$ ,  $20.47^\circ \text{ E}$ ,  $132 \text{ m}$ ), Serbia. The Belgrade climate is moderate continental, with the average daytime temperature of  $11.7^\circ \text{ C}$  and the average amount of precipitation of about  $700 \text{ mm/year}$ . The

highest daily temperatures, above  $30\text{-}35^\circ \text{ C}$ , are registered in July and August.

Aerosol samples were collected on filter papers (FILTRAK/Whatman 41/DDR,  $15 \text{ cm}$  diameter, with  $80\%$  dust retention efficiency) by constant flow rate samplers (average air flow  $20 \text{ m}^3/\text{h}$ , average daily volume  $600 \text{ m}^3$ ). The samples were ashed at temperatures below  $400^\circ \text{ C}$  and a composite monthly sample was formed from daily filters (average volume  $15 \cdot 10^3 \text{ m}^3$ ). The composite samples were measured in small metallic containers.

The activities of the radionuclides were determined on three Canberra HPGe detectors by standard gamma spectrometry. An  $n$ -type reverse coaxial detector (named Detector 2), with  $20\%$  relative efficiency, has a high efficiency in low-energy part of the spectrum. The other two  $p$ -type detectors (named Detector 1 and Detector 3) have relative efficiencies of  $18\%$  and  $50\%$ . All detectors have a lead shielding that provides low background thus enabling low-activity measurements. Detectors 2 and 3 have a commercial lead shielding with an inner copper layer of  $3 \text{ mm}$ . The shielding for Detector 1 was custom built, and it consists of  $10 \text{ cm} \cdot 10 \text{ cm} \cdot 10 \text{ cm}$  lead cubes with an inner layer of  $10 \text{ mm}$  iron and  $3 \text{ mm}$  copper.

Energy calibration was performed with a set of standard point sources (Coffret d'etalon gamma ECGS-2, Sacle, France, containing  $^{133}\text{Ba}$ ,  $^{57}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$ , with activities in the range of  $10^3\text{-}10^4 \text{ Bq}$ , 25. 11. 1987). Full energy peak efficiency was determined with the IAEA-083 (AIR4) simulated air filter (spiked with solution of  $^{60}\text{Co}$ :  $2160 \text{ Bq/filter}$ ,  $^{133}\text{Ba}$ :  $846 \text{ Bq/filter}$ ,  $^{137}\text{Cs}$ :  $1182 \text{ Bq/filter}$  and  $^{210}\text{Pb}$ :  $151 \text{ Bq/filter}$ , uncertainty  $5\%$ , 1. 1. 86) for measurements conducted during 2004-2008. During 2008-2012, efficiency calibration was performed using a secondary reference material (aerosol dust matrix) produced from the radioactive solution 9031-OL-116/08 and 9031-OL-427/12 issued by the Czech Metrological Institute, Inspectorate for Ionizing Radiation, which contained  $^{210}\text{Pb}$ ,  $^{241}\text{Am}$ ,  $^{57}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{113}\text{Sn}$ ,  $^{139}\text{Ce}$ ,  $^{85}\text{Sr}$ ,  $^{109}\text{Cd}$ ,  $^{88}\text{Y}$ , and  $^{203}\text{Hg}$ .

Coincidence summing correction was not performed for calibration curves. The efficiency at  $46 \text{ keV}$  was derived directly from the calibration source measurement:  $\varepsilon = 2\text{-}3\%$  for Detectors 1 and 3, and  $\varepsilon = 12\%$  for Detector 2. In the case of  $^7\text{Be}$ , the efficiency was derived from the calibration curve without coincidence correction. Our latest investigation using EFTRAN coincidence correction software [36] showed that, within the uncertainty budget, the uncorrected efficiency was the same as the corrected efficiency for the  $477 \text{ keV}$  energy.

Uncertainty budget  $\sigma_{\text{ef}}$  was calculated as follows

$$\sigma_{\text{ef}} = \sqrt{\left(\frac{\Delta A_s}{A_s}\right)^2 + \left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta P_\gamma}{P_\gamma}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}^{1/2} = \sqrt{(4\%)^2 + (1\%)^2} \quad (1)$$

where  $\Delta A_s$  is the activity uncertainty in radioactive solution, as given by the manufacturer,  $N$  – the uncertainty of the peak area,  $P_\gamma$  – the uncertainty of the emission probability at given energy,  $t$  – the uncertainty of the measurement time (negligible), 4% – the estimated uncertainty introduced *via* preparation of the secondary reference material, and 1% – the estimated uncertainty introduced *via* fitting of the experimental points with the calibration curve.

A typical background  $^{210}\text{Pb}$  spectrum contains 150-250 counts for a measurement period of 60,000 s (0.0025 cps-0.004 cps) with a standard uncertainty of 5%, while a typical air filter spectrum contains about 1000 counts with standard uncertainty of 3% for the same measurement period.

The activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  were determined at gamma energies of 477 keV and 46 keV, respectively. The minimum detectable concentrations were derived using the lower limit of detection  $LLD$ , as  $LLD = k^2 \cdot 2LC$ , where  $k$  is the coefficient of normal distribution, and  $LC$  is the critical level depending on background photo peak counts [37]. The value of the  $k$  factor in the  $LLD$  formula is 1.645 [38], and by definition is not the same as the level of confidence defined in the uncertainty. The minimum detectable concentration  $MDC$  was calculated as  $MDC = LLD / (p \cdot \epsilon \cdot t \cdot V \cdot \epsilon_f)$ , where  $p$  is the probability of gamma emission,  $\epsilon$  – the efficiency,  $t$  – the counting interval,  $V$  – the volume of the air sample, and  $\epsilon_f$  – the filter paper efficiency.

In our measurements, the counting time intervals ranged from 60,000 s to 250,000 s. The minimum detectable concentrations were taken as  $10 \mu\text{Bq}/\text{m}^3$  for  $^7\text{Be}$  and  $20 \mu\text{Bq}/\text{m}^3$  for  $^{210}\text{Pb}$ , which represent the average  $MDC$  for our system. For example, with a typical  $^{210}\text{Pb}$  background count of 150-200 (as previously mentioned), the calculated  $LLD$  is 60-76 counts, which corresponds to  $MDC$  of 14-80  $\mu\text{Bq}/\text{m}^3$ .

The results were statistically analyzed by Genie 2000 program. Measurement uncertainty is expressed as an expanded measurement uncertainty for the factor

$k=2$  which corresponds to a normal distribution with a confidence level of 95%.

The sunspot data were obtained from the SIDC-team (World Data Center for the sun-spot index, Royal Observatory of Belgium, Monthly Report on the International Sunspot Number, online catalogue of the sunspot index: <http://www.sidc.be/sunspot-data/2003-2012>). Precipitation data for 2004-2009 for the Institute site were provided by the Department of Meteorology of the Institute. The meteorological data for the City site were obtained from the European Climate Assessment & Dataset (ECA&D) [39] and the Republic Hydrometeorological Service of Serbia.

## RESULTS AND DISCUSSION

### Time series of $^7\text{Be}$ and $^{210}\text{Pb}$ activities

The activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in two locations (Institute and City) over the 2004-2012 period are given in figs. 1 and 2. The differences in the  $^7\text{Be}$  data arrays of monthly activities measured in the two locations were not significant (Student's T-test significance 0.102), unlike the  $^{210}\text{Pb}$  activities which differed significantly (Student's T-test significance 0.0052).

To investigate a relation between the activities time series, Pearson's linear correlation coefficients ( $r$ ) were calculated, and their statistical significance was tested at the 0.05 significance level. At each measuring site,  $r$  showed a positive correlation between the radionuclides, 0.35 and 0.30 for Institute and City, respectively, which is statistically significant at the 0.05 level. This result is in agreement with a monitoring study showing mutually correlated activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in an urban area [40]. On the other hand, the correlation of the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities across the sites was poor ( $r=0.18$ ) and not significant at the 0.05 level, which may indicate that the radionuclides' activities in surface air are to some extent influenced by the local meteorological conditions.

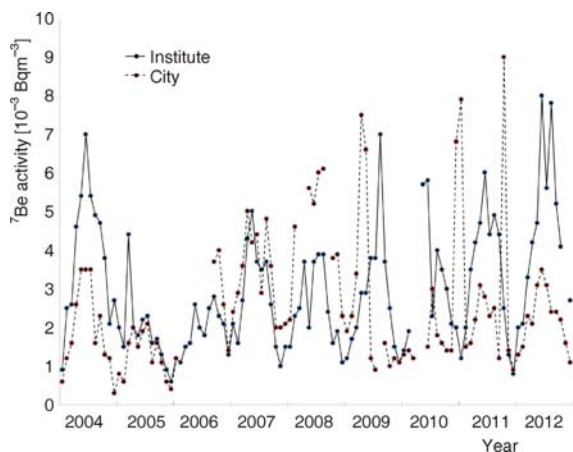


Figure 1.  $^7\text{Be}$  activities in Belgrade, 2004-2012 (the line interruptions mark missing data)

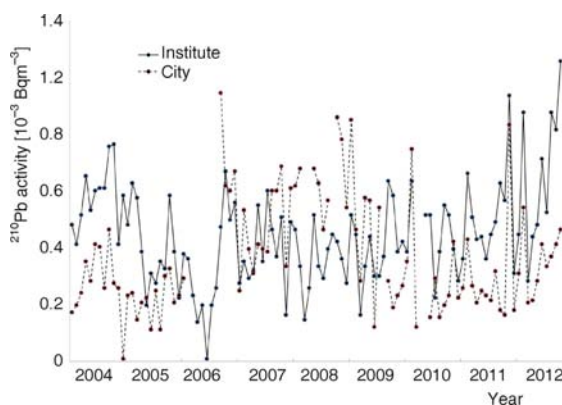


Figure 2.  $^{210}\text{Pb}$  activities in two locations in Belgrade, 2004-2012 (the key as in fig. 1)

The seasonal patterns for  $^7\text{Be}$  and  $^{210}\text{Pb}$  (figs. 3 and 4, respectively) are in agreement with previous studies [16, 21]. Comparison of the overall means for the investigated period (figs. 3 and 4) with the means given in [16, 21] offers no evidence for temporal trends in the radionuclides' activities. This result agrees with the finding that during 1972-2003 there were no changes in the  $^7\text{Be}$  activities in Europe [41]. However, a  $^7\text{Be}$  decrease in surface air was noted over the 1970-1997 period [42], which the authors partly explained by a possibility that an increased precipitation scavenged this radionuclide from air masses before they reached the ground level where they were sampled.

### Influence of solar activity on the $^7\text{Be}$ concentration

The mean  $^7\text{Be}$  activities and number of sunspots in each season were calculated (March, April, and May in the spring season; June, July, and August in the summer season; September, October, and November in the autumn season; and December, January, and February

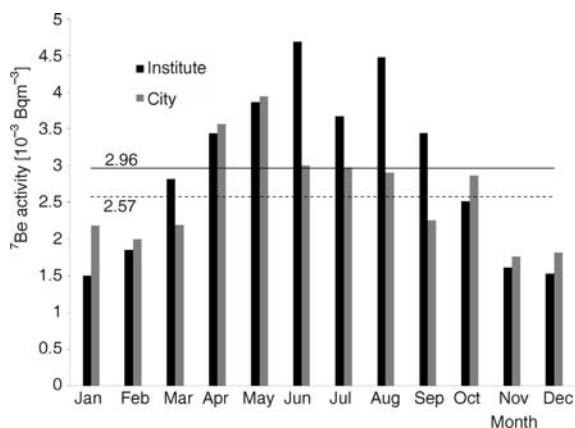


Figure 3. Monthly means of  $^7\text{Be}$  activities (the full and dotted lines denote the overall means for the Institute and City site, respectively)

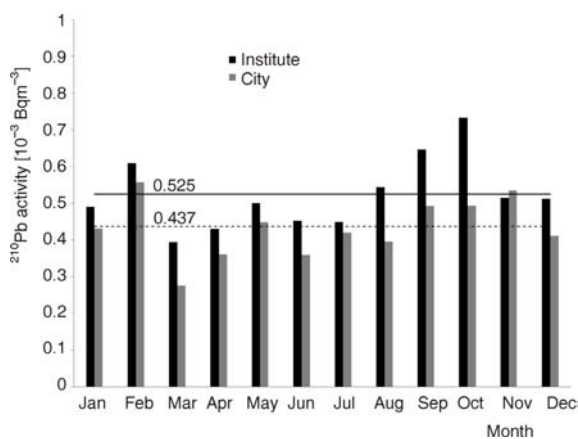


Figure 4. Monthly means of  $^{210}\text{Pb}$  activities (the key as in fig. 3)

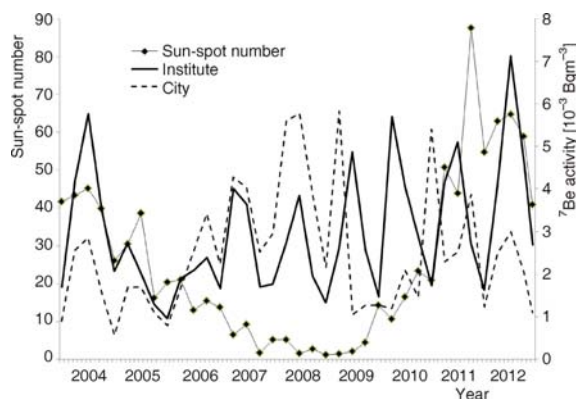


Figure 5. Seasonal activities of  $^7\text{Be}$  and seasonal sun-spot numbers

in the winter season) and their relation was then examined. At the Institute site (fig. 5) they were negatively correlated ( $r = -0.56$ ), but the correlation was not significant at the 0.05 level. The reported correlation coefficients in studies [7, 8, 41] which covered longer periods of time than our analysis, showed a somewhat stronger anti-correlation. Further, a phase-shift of a few months was found in the correlation between the sun-spot number and the surface  $^7\text{Be}$  activity [7, 41], corresponding to the time necessary for the changes in solar wind to reflect on the amount of the radionuclide in surface air. In our data, however, the influence of the vertical transport time was not evident as  $r$  for Institute ranged between  $-0.55$  and  $-0.61$  when time-lag of 1-6 months was included in the calculations, but the correlation coefficients were not statistically significant. At the City site, the calculated  $r$  was in the range ( $-0.29$ ,  $-0.20$ ) and was not significant at the 0.05 level.

### $^7\text{Be}$ and $^{210}\text{Pb}$ activities and their correlations with meteorological variables

The correlations of the radionuclides' activities with the local meteorological variables are presented in tab. 1. The precipitation measurements were the sole data available for the Institute site. Even though this dataset covered only the 2004-2009 period (shorter than the 2004-2012 data available at the City site), during that period, the precipitation data at Institute and City were well correlated ( $r = 0.76$ ) and the correlation was significant.

Over the investigated periods, there was no statistically significant correlation found between the radionuclides' activities and precipitation (tab. 1). This result agrees well with other multi-year studies [8, 30], contrasting a negative correlation ( $r = -0.594$ ) between the  $^7\text{Be}$  activity and rainfall found in a study encompassing a period of five years [12].

To further investigate the relation between the radionuclides' activities and precipitation, the total monthly precipitation data were grouped into bins of

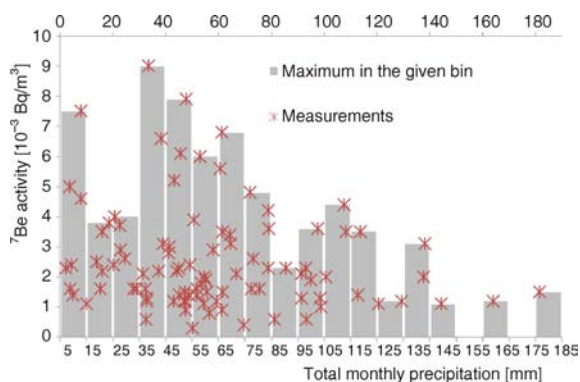
**Table 1. Pearson's linear correlation coefficients (*r*) for the <sup>7</sup>Be and <sup>210</sup>Pb activities and precipitation, temperature, relative humidity, cloud cover, sunshine hours and atmospheric pressure; statistically significant correlation is given in bold**

	Precipitation Institute	Precipitation City	Temperature	Relative humidity	Cloud cover	Sunshine hours	Pressure
<sup>7</sup> Be	-0.04	-0.20	<b>0.29</b>	-0.43	-0.37	<b>0.38</b>	-0.05
<sup>210</sup> Pb	-0.25	-0.19	-0.05	-0.01	-0.12	0.02	<b>0.29</b>

different widths. Figure 6 shows the <sup>7</sup>Be activity (City site) as a function of the total monthly precipitation (marked by asterisks) as well as 19 bins of total monthly precipitation (the minimum value 0 mm, the maximum 190 mm, the bin width 10 mm) with the maximum <sup>7</sup>Be activity that was measured in each of the bins. A negative correlation between these maximum activities and the centre values of the bins ( $r = -0.76$ ) were found. Similarly,  $r = -0.82$  was obtained for the maximum <sup>210</sup>Pb activities corresponding to the binned total monthly precipitation data.

A different choice of the bin width of the precipitation data resulted in different values of the *r* coefficient. The wider the bin (*i. e.*, fewer number of precipitation bins), the stronger linear negative correlation was obtained. For example, correlation with the <sup>7</sup>Be activity increased to  $r = -0.94$  when the number of precipitation bins was reduced to 10 (the bin width 20 mm), whereas a choice of 38 bins (the bin width 5 mm) decreased the correlation to  $r = -0.58$ . However, the statistical test showed that these correlations were not significant at the 0.05 level.

The relationship between the maximum activity and the binned precipitation data (fig. 6) indicates two to three possible modes of radionuclide washout from the atmosphere. The first mode encompasses very dry months, with the total precipitation lower than 30 mm, and the majority of the <sup>7</sup>Be activities below 4 mBq/m<sup>3</sup>. Nearly a half of the months under the 30 mm total precipitation limit were in autumn, which is a dry season in Belgrade. Within this mode, there were three exceptions when the <sup>7</sup>Be activities were higher than 4 mBq/m<sup>3</sup>: April 2007, February 2008, and April 2009. The corresponding <sup>7</sup>Be activities were 5.0 mBq/m<sup>3</sup>, 4.6 mBq/m<sup>3</sup>, and 7.5 mBq/m<sup>3</sup>, respectively, and they all exceeded the seasonal means (fig. 3) by at least 50%. Interestingly,

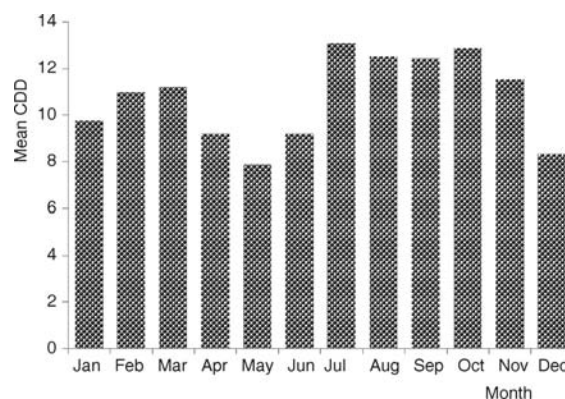


**Figure 6. <sup>7</sup>Be activities and total monthly precipitation at the City site**

only April 2007 was preceded by a dry month, the fact which could explain the observed high activity as a consequence of the radionuclide accumulation during the dry spell.

Another meteorological index, which serves as an indicator of the precipitation extremes, shed some more light on the radionuclide concentration increase during dry episodes. The maximum number of consecutive dry days (hereinafter, CDD) in each of the anomalous three months, as well as in their preceding months, was higher than the corresponding monthly mean (fig. 7): in April 2007 CDD = 17 and in March 2007 CDD = 16; in February 2008 CDD = 22 and in January 2008 CDD = 10; in April 2009 CDD = 12 unlike March 2009 when CDD = 5 was lower than the monthly mean. Over the March-April period in 2007, there were actually 32 consecutive days with no precipitation, with the exception of two days when precipitation was 0.1 mm and 1.0 mm. This very long dry period most likely contributed to an atypically high <sup>7</sup>Be activity in April 2007. Similarly, although in February 2008 the 22 consecutive dry days did not follow on from the previous month, the increased <sup>7</sup>Be concentration was at least partly influenced by the lack of atmospheric wet deposition. The April 2009 episode, on the other hand, stands out as an exception to this explanation.

The second mode in the activity-precipitation relation (fig. 6) is characterized by a decrease in the maximum activity as precipitation increases. The third mode, on the other hand, with the total monthly precipitation greater than 120 mm (only seven data points) may imply a saturation whereby an increase in precipitation does not result in further decrease of the radionuclide activity. The monthly precipitation accu-



**Figure 7. Monthly means of the number of consecutive dry days in a month**

mulations over 120 mm were mainly connected with summer episodes, and could be considered as precipitation extremes. Intensive convective clouds followed by an intensive precipitation, are probably very effective in washing out the radionuclides attached to aerosols. In these types of clouds, especially when they are well developed, precipitation and wide range of different hydrometeors exist throughout the tropospheric layer, which is thus thoroughly cleansed. All of the seven months in this regime were characterized by at least four days of heavy precipitation (greater than 10 mm), 1-3 days of very heavy precipitation (greater than 20 mm) and CDD between 5 and 7, in each case less than the corresponding monthly mean.

The above analysis indicated that there could be a positive correlation between the  $^7\text{Be}$  activity and the number of consecutive dry days, but the calculated correlation coefficient did not show a relationship between these variables ( $r = 0.03$ ). Therefore, the impact of CDD on the radionuclide activity could not be quantified, even though its role in enabling the radionuclide accumulation during dry periods seems significant. In extremely wet months, on the other hand, CDD might not have been long enough to allow replenishing surface air with  $^7\text{Be}$  from higher altitudes.

A similar pattern, with the pronounced three different modes, was seen in the  $^{210}\text{Pb}$  activity and binned precipitation relation. These results are in good agreement with three different modes in a relationship of the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities with precipitation noted by [10].

Influence of other meteorological variables (temperature, relative humidity, cloud cover, sunshine hours, and atmospheric pressure) on the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities was also examined (tab. 1). Statistically significant correlation was found only for the  $^7\text{Be}$  activity with temperature ( $r = 0.29$ ) and sunshine hours ( $r = 0.38$ ), and for the  $^{210}\text{Pb}$  activity with pressure ( $r = 0.29$ ). The increase in the  $^{210}\text{Pb}$  activity, whose primary source is in the lower-most air (at the very top of soil), with the rise in atmospheric pressure could be explained by a hindered upward lift – a high pressure situation, accompanied by air subsidence, can contribute, especially when surface winds are low, to shallower planetary boundary layer thus causing an increased surface concentration of any well mixed tracer inside the layer.

Our correlation results are in partial agreement with the findings of [8, 12, 30, 43]. For example, the study of [8], encompassing the longest examination period (1998-2009), showed: (1) a positive correlation between the radionuclides' activities and temperature, which is in agreement with our statistically significant correlation between  $^7\text{Be}$  and temperature; (2) no correlation between the  $^7\text{Be}$  activity and relative humidity, but positive correlation between  $^{210}\text{Pb}$  and relative humidity, which agrees with our finding for  $^7\text{Be}$ ; and (3) no correlation with atmospheric pressure for either of

the radionuclides. This lack of correlation between the  $^{210}\text{Pb}$  activity and atmospheric pressure is dissimilar to our result ( $r = 0.29$  in tab. 1). The explanation may lie in the fact that the air masses examined in [8] were partly of maritime origin (with an increased relative humidity), and therefore were not as rich in  $^{210}\text{Pb}$  as continental air masses [25]. The same argument could explain the positive correlation between relative humidity and the  $^{210}\text{Pb}$  activity found in [8], but not in our analysis.

Our data offered some insight into the impact of the investigated variables on the concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in surface air. The temporal resolution of the radionuclides' behaviour was limited by the availability of the aerosol samples (a composite monthly sample), thus allowing only changes on a seasonal scale to be captured. Further, the same sets of meteorological variables were not available at both of the measuring locations, whereby only a partial analysis on the influence of the local climate (namely, the effect of precipitation) could be performed. Another drawback of our study was the inability to examine the role of horizontal transport, as our monthly data only provided an averaged and stationary picture of a more complex interplay between the production, transport and removal of the radionuclides in the atmosphere. Nevertheless, the obtained results shed more light on the behavior of the radionuclides in the atmosphere and the governing mechanisms.

## CONCLUSIONS

During 2004-2012, the activities of  $^7\text{Be}$  and  $^{210}\text{Pb}$  were determined in composite monthly aerosol samples collected from surface air in two locations in Belgrade. The radionuclides' activities, their mean values and seasonal patterns were in good agreement with the previous studies in the region. A statistically significant correlation was found between the radionuclides at each of the measuring sites. In contrast, no significant correlation was obtained for the radionuclides' activities across the two locations, indicating a notable impact of the local meteorological conditions on the activities in surface air.

A strong negative correlation ( $r = -0.56$ ), however not statistically significant, between the cosmogenic  $^7\text{Be}$  and number of sun-spots was found at the Institute site, while no correlation was found at the City site.

The role of precipitation in the abundance of the radionuclides was investigated in greater detail. At both of the sites, there was no correlation between the activities and precipitation. For both of the radionuclides, three distinct modes of radionuclides' washout from the atmosphere were found. Within the temperate range of the total monthly precipitation, the maximum activities decreased with precipitation. The other two modes describe dry and very wet months. Longer periods of dry weather

seemed to enable the radionuclides' accumulation in the atmosphere, but no correlation was found between the number of consecutive dry days and the activities.

The influence of temperature, relative humidity, cloud cover, sunshine hours, and atmospheric pressure on the  $^7\text{Be}$  and  $^{210}\text{Pb}$  activities was also examined. Statistically significant correlation was found only for the  $^7\text{Be}$  activity with temperature and sunshine hours, and for the  $^{210}\text{Pb}$  activity with atmospheric pressure.

#### ACKNOWLEDGEMENTS

This paper was realized as a part of the project "Studying climate change and its influence on the environment: impacts, adaptation and mitigation" (43007) financed by the Ministry of Education, Science and Technological Development of the Republic of Serbia within the framework of integrated and interdisciplinary research for the period 2011-2014. The authors would like to thank the Republic Hydrometeorological Service of Serbia for the provision of meteorological data.

#### AUTHOR CONTRIBUTIONS

Experiments were carried out by D. Todorović and J. Nikolić, and theoretical analysis was performed by J. Ajtić and V. Djurdjević. All of the authors analysed and discussed the results. The manuscript was written by J. Ajtić and V. Djurdjević, and the figures were prepared by J. Ajtić.

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Received on July 22, 2013

Accepted on December 24, 2013

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**ВИШЕГОДИШЊА СТУДИЈА РАДИОАКТИВНОСТИ  
У ПРИЗЕМНОМ СЛОЈУ АТМОСФЕРЕ У РЕЛАЦИЈИ  
СА КЛИМАТСКИМ ПАРАМЕТРИМА У БЕОГРАДУ, СРБИЈА**

Активности  $^7\text{Be}$  и  $^{210}\text{Pb}$  мерене су током 2004-2012, у приземном слоју атмосфере на две локације у Београду. Месечне специфичне активности одређене су стандардном методом спектрометрије гама зрачења на HPGe детекторима. Активности радионуклида показују сезонски карактер:  $^7\text{Be}$  има максимум током пролећа и лета, а минимум зими, док  $^{210}\text{Pb}$  има два максимума, током јесени и касне зиме. На обе локације, средње месечне активности  $^7\text{Be}$  су мање од  $9 \text{ mBq/m}^3$ , док су за  $^{210}\text{Pb}$  мање од  $1.3 \text{ mBq/m}^3$ . Између активности  $^7\text{Be}$  и броја сунчевих пега није нађена статистички значајна корелација. Релације између активности ових радионуклида и климатских параметара (количина падавина, температура, релативна влажност, облачност, број сунчаних сати и атмосферски притисак) такође су испитане, али су значајне корелације добијене само за активност  $^7\text{Be}$  са температуром и бројем сунчаних сати, односно за активност  $^{210}\text{Pb}$  са атмосферским притиском. На основу везе између максималне активности по интервалима количине падавина, могу се разликовати три режима спирања радионуклида из атмосфере. Током сушних периода, акумулација радионуклида у атмосфери узрокује њихову повишену активност, али корелација између активности и броја застопних сувих дана није добијена.

*Кључне речи: радиоактивност,  $^7\text{Be}$ ,  $^{210}\text{Pb}$ , атмосфера, број сунчевих пега, климатски параметар*