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CONCENTRATION MEASUREMENTS OF ^{7}Be AND ^{137}Cs IN GROUND LEVEL AIR IN THE BELGRADE CITY AREA

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Concentrations of ^{7}Be and ^{137}Cs in ground level air in the city area (Belgrade, central Serbia) were determined in the period from 1991-1996. The average monthly concentrations of ^{7}Be in ground level air were in the range of 2-7 mBq/m^3 with pronounced one or two maxima in summer or early fall and a minimum in winter. The average air concentrations for ^{137}Cs were from 0.5-8.5 x 10-5 Bq/m^3, with a spread maximum in the spring-summer period and a pronounced maximum during the winter. A general increase in ^{7}Be and ^{137}Cs concentrations during 1993 was recorded. The maximum seasonal indices were 1.3 for ^{7}Be (summers) and 2.7 (late springs and early summers) and 3.4 (winters) for ^{137}Cs. No correlation with the amount of precipitation and ^{137}Cs concentrations in air was determined, while the washout effect of rainfalls seems to be more closely related with variations in ^{7}Be concentrations.

INTRODUCTION

Long-term measurements of ^{7}Be and other cosmogenic and atmospheric radionuclides provide a tool to study large-scale atmospheric processes and to compare the environmental impact of radioactivity from man-made sources with natural ones, while seasonal variations of ^{137}Cs concentrations in ground air indicate the migrations from stratosphere to troposphere, and eventually point to the changes in the global ecosystem (Gustafson et al. 1981; Agelao et al. 1984; Baeza et al. 1996; Papastefanou and Joannidou 1995; Rosner et al. 1996).

Continuous monitoring of ^{7}Be concentrations in ground level air at middle latitudes in the last decade indicated the average values of ^{7}Be concentration to be up to 10 mBq/m^3, with a general increase in the ^{7}Be concentrations recorded during 1993 (Larsen et al. 1995; Cannizzaro et al. 1995). Variations in the annual mean ^{7}Be concentrations in the period were attributed mainly to the changes in the atmospheric production rate, while seasonal maxima of ^{7}Be concentrations were considered to be primarily due to the strong stratosphere to troposphere exchanges in summers,

typical of these latitudes (Todorovic 1997; Durana et al. 1996; Hartwig 1996).

Concentrations of ^{137}Cs in ground level air in the same period were of the order of $\mu Bq/m^3$, with one or two maxima, generaly during summers and winters (Larsen et al. 1995; Cannizzaro 1995). Some authors attribute ^{137}Cs winter maximums to the inversion weather conditions (Bunzl et al. 1995), while others tend to the conclusion that in the 1990s, the main source of ^{137}Cs is surface air resuspension of soil dust from the Chernobyl fallout, while the stratospheric contribution is much less significant (Fresnel et al. 1996). The calculations of the ^{137}Cs summer max/min ratio also indicates a slow decrease of stratospheric ^{137}Cs originated by Chernobyl (Bunzl 1995; Hartwig 1996).

In Yugoslavia, there are 30 years of data on the monitoring of ^{137}Cs concentrations in ground level air, but continuous measurements of ^{7}Be concentrations started only a few years ago (Bauman 1967; Todorovic et al. 1996, 1997).

This paper presents data on the concentration measurements of ^{7}Be and ^{137}Cs in the ground level air at a meteorological station, Usek, in the region of the Institute Vinca in the Belgrade City area, in the period from 1991-1996. Measurements were performed within a project investigating stratosphere to troposphere migration and exchange processes (Todorovic 1997).

MATERIALS AND METHOD

Air samples were collected daily at the meteorological station Usek, 2 km from the Institute "Vinca" in the direction of wind towards the city (Belgrade, central Serbia). Samples were collected on filter paper (diameter 15 cm, filter relative efficiency for freely deposited dust 80%) using constant flow rate samplers (average air flow 20 m^3/h, average daily air volume 600 m^3) on a collector site placed 1 m above the ground. Filters were changed daily. Daily samples were ashed at temperatures below 400 °C to improve the detection efficiency and minimize the minimum detectable concentrations. A monthly composite sample consisting of about 30 daily filters was formed in planchets (average volume 15 x 10^3 m^3).

The activities of ^{7}Be and ^{137}Cs in air (Bq/m^3) were determined on an HPGe detector (vertical, coaxial type, ORTEC, relative efficiency 23%) by standard gamma spectrometry. The detector was placed in a shielding cage (45 x 45 x 45 cm) of Pb bricks (width 10 cm) with layers of Cu (3 mm) and Fe (6 cm) on the inner side. Energy calibration was performed with a set of standard point sources (CEFFRET d, etalon gamma ECGS-2, Sacle, France) containing ^{133}Ba, ^{57}Co, ^{60}Co, and ^{137}Cs (mean activity 10^2 Bq, overall uncertainty 3%). Geometric efficiency was determined by a reference aerosol sample (ZND 1989).

Counting time intervals ranged from 150 000 - 250 000 s. The background spectrum (integral mean counts 1,7 imp/s) was recorded regularly immediately after or before the sample counting. The total standard error of the method (calculated as the sum of relative errors in geometric efficiency estimation, photo peak counts estimation, sample volume determination, etc.) was estimated below 15%.

Minimum detectable concentrations (derived from the lower limit of detection defined as $LLD = k^2 \pm 2LC$, where k is a coefficient of normal distribution corresponding to confidence level 95% and LC is the critical level depending on the background photo peak counts) for ^{137}Cs and ^{7}Be were 1.8 $\mu Bq/m^3$ and 50 $\mu Bq/m^3$, respectively.

The data were statistically analysed on an IBM/PS2 with the SPECTRAN-AT programme.

Precipitation data for the period were obtained from the Department of Meteorology, Institute of Nuclear Sciences "Vinca" (DM-INS Reports 1991-1996).

RESULTS AND DISCUSSION

The average monthly concentrations of ^{137}Cs and ^{7}Be in ground level air in the Belgrade City area in the period from 1991-1996 are presented in Fig. 1 and Fig. 2, respectively. The average monthly precipitation values for the same period are presented in Fig. 3. The standard error of the method was less than 15%.

The average monthly ^{7}Be concentrations in ground level air in the period were in the range 2-7 mBq/m^3, with pronounced one or two maxima in summer or early fall and a minimum in winter. The overall average concentration (± standard deviation) for the whole period was 4.04 ± 1.65 mBq/m^3. The data correspond with values measured on the middle latitudes reported in literature. Also, an increase of about 20% in the ^{7}Be concentration was recorded in 1993. The average concentration of ^{137}Cs in ground level air in the same period was in the range 0.5-8.5 x 10-5 Bq/m^3, with the overall average concentration (± standard deviation) for the whole period of (2.46 ± 1.04) x 10-5 Bq/m^3. A pattern with a maximum in the spring-summer period and another pronounced maximum during winter was observed.

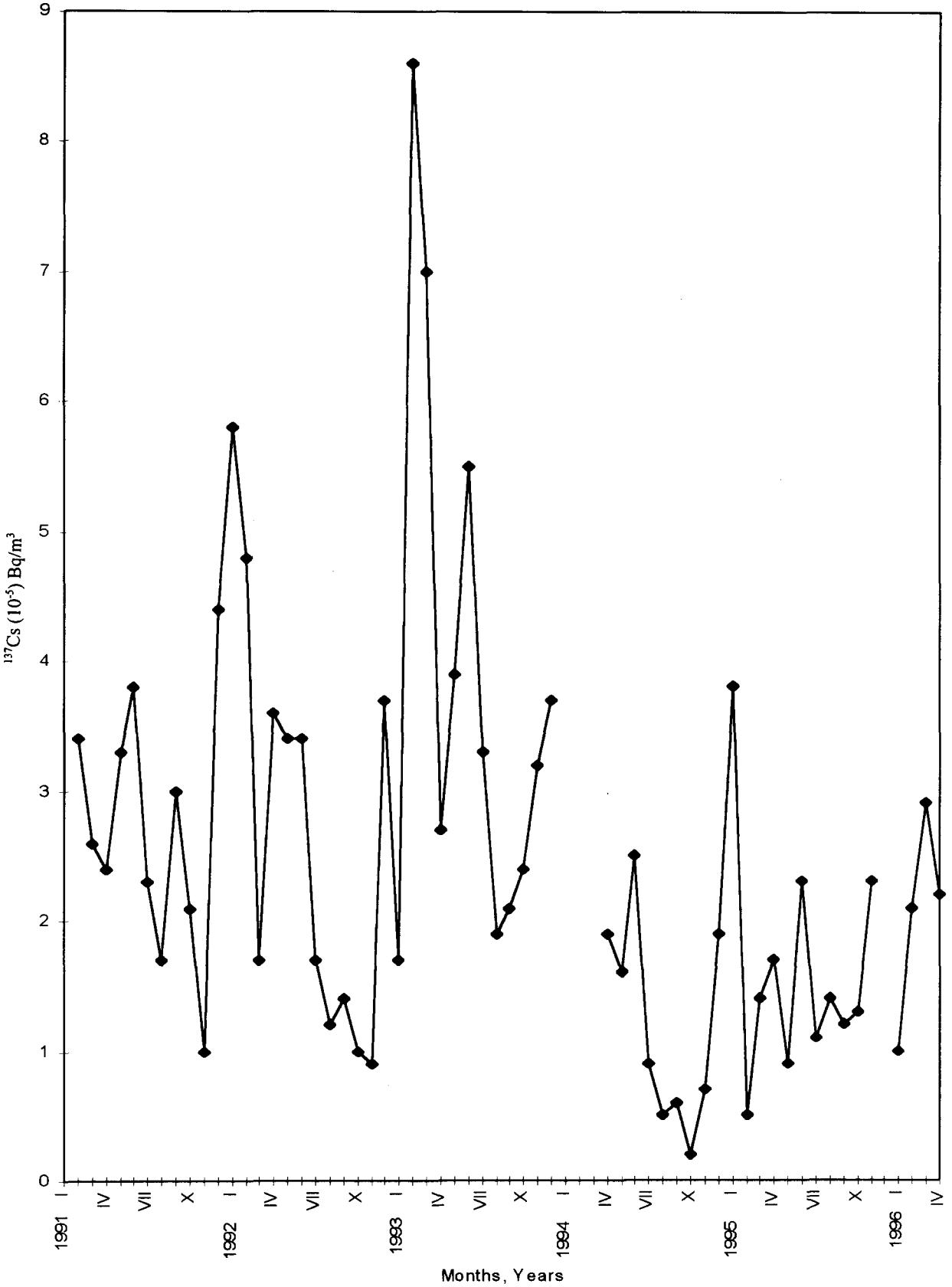

Fig. 1. Average monthly ^{137}Cs concentrations in ground level air in the Belgrade City area from 1991-1996.

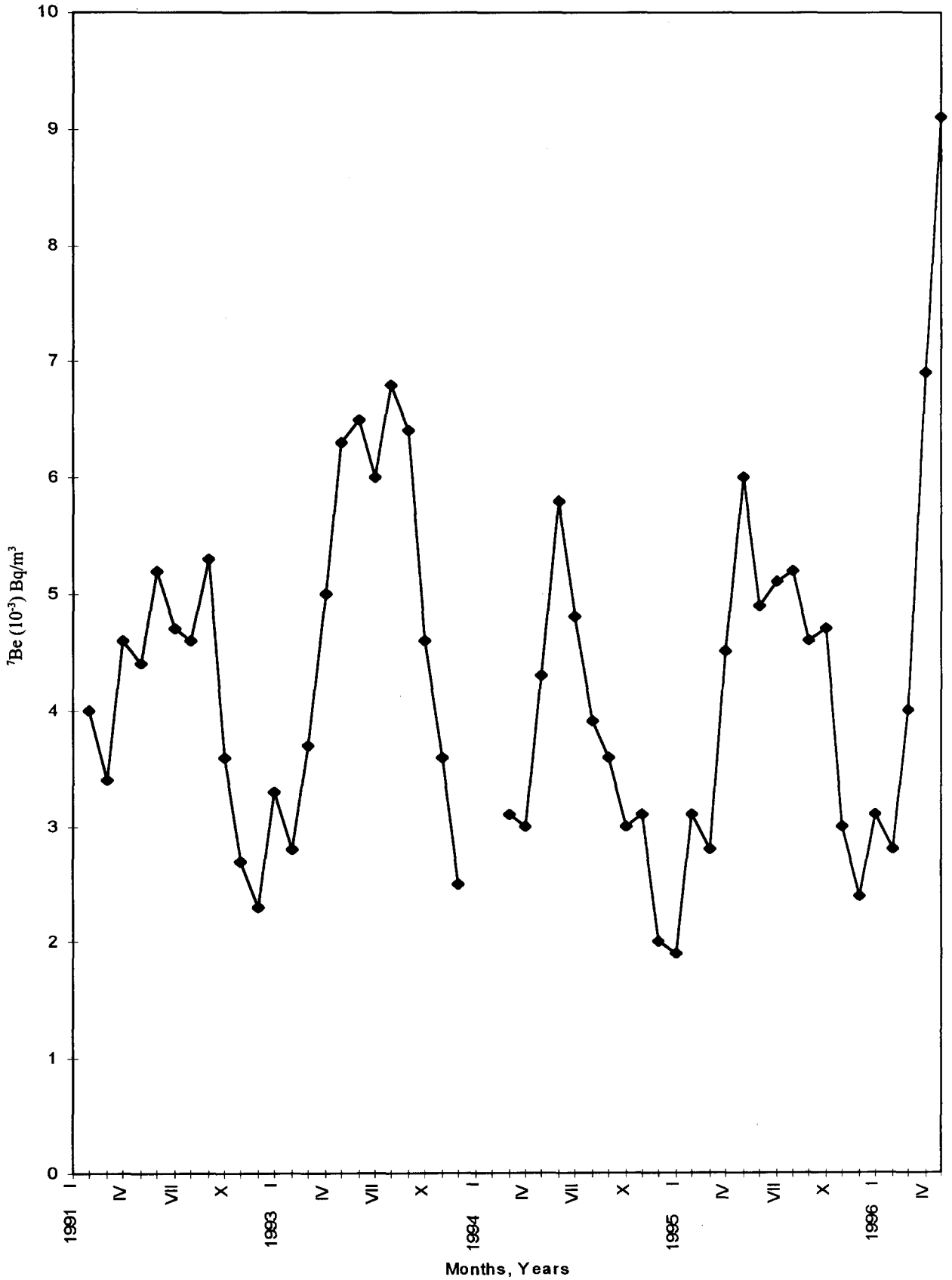

Fig. 2. Average monthly ^{7}Be concentrations in ground level air in the Belgrade City area from 1991-1996.

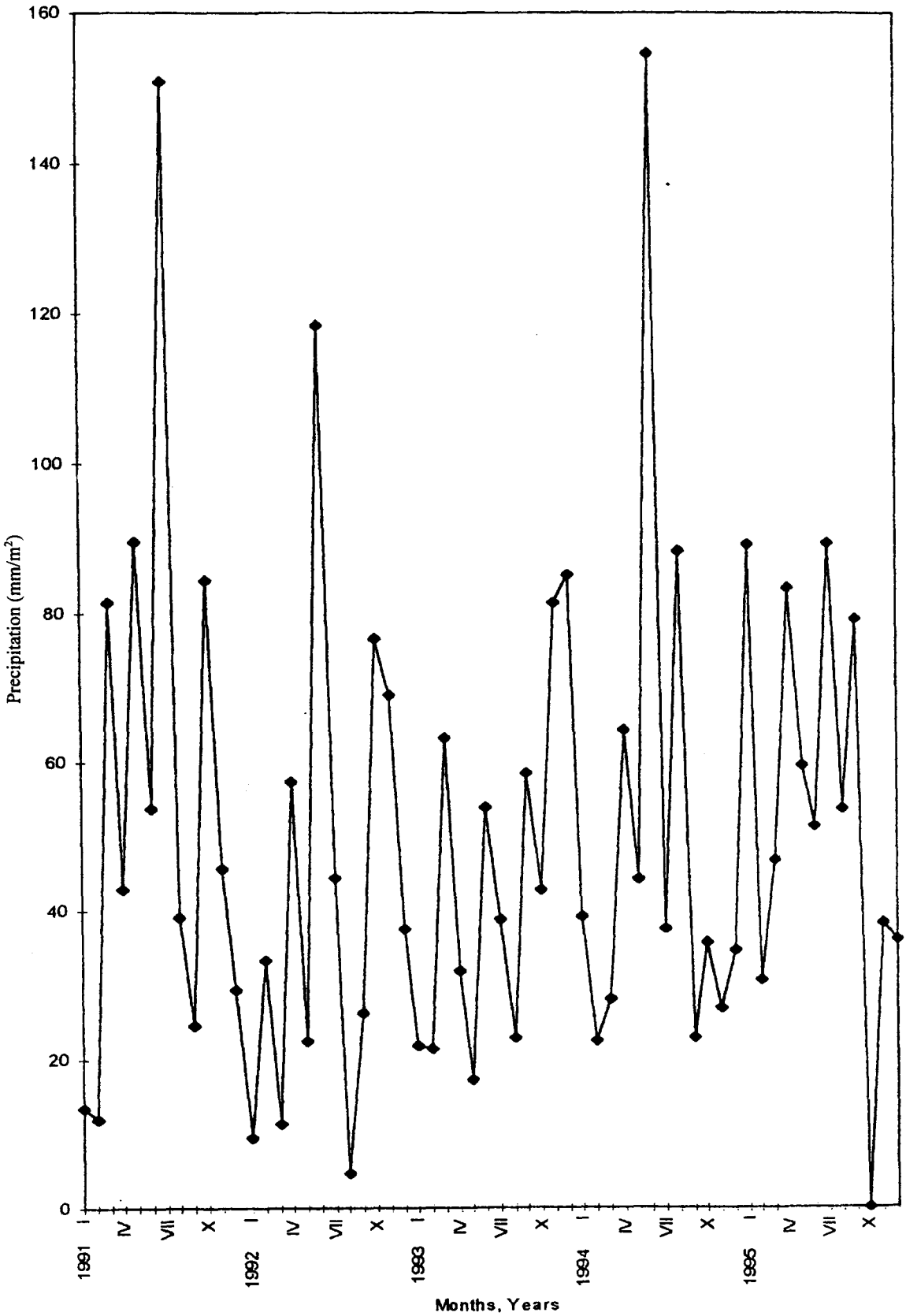

Fig. 3. Average monthly precipitation values in the Belgrade City area from 1991-1996.

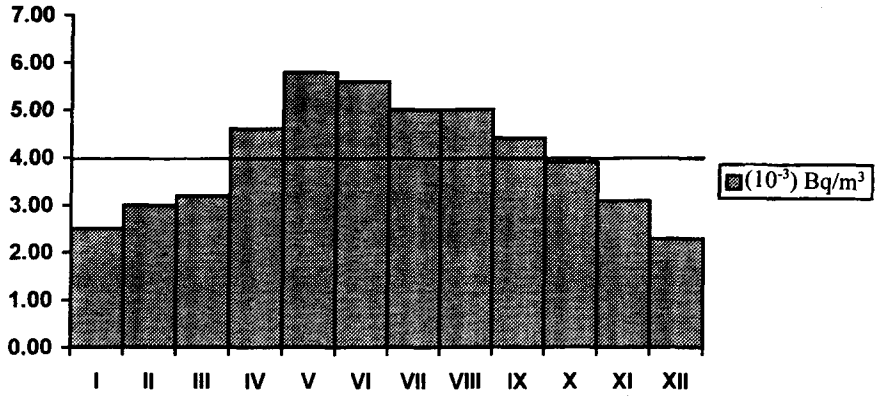

Fig. 4. Histogram of the means of the average monthly ^{7}Be concentrations in the period 1991-1996 (the horizontal line is the overall mean for the period).

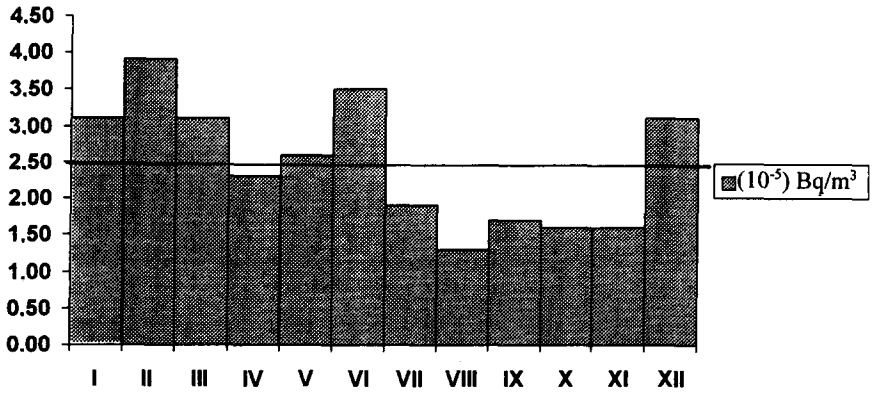

Fig. 5. Histogram of the means of the average monthly ^{137}Cs concentrations in the period 1991-1996 (the horizontal line is the overall mean for the period).

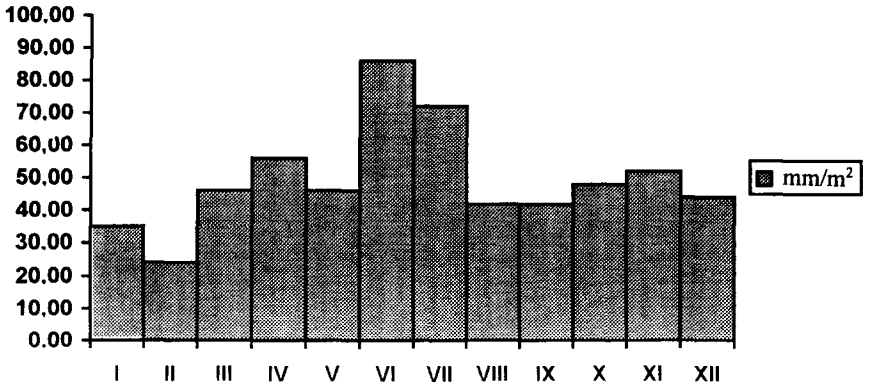

Fig. 6. Histogram of the means of the average monthly amounts of precipitation in the period 1991-1996.

To provide statistical evidence for the observed seasonal trends in ^{7}Be and ^{137}Cs concentrations, the means of the average monthly concentrations for the period, the seasonal indices, the average quarterly seasonal indices for the period, and the maximum/ minimum ratios were calculated.

The means of the average monthly concentrations of ^{7}Be and ^{137}Cs in the ground level air in the period 1991-1996 are presented in Figs. 4 and 5, respectively. The means of the average monthly precipitation values are presented in Fig. 6 for comparison. The standard deviation for the calculated means was less than 20%.

Table 1. Seasonal indices for ^{7}Be and ^{137}Cs from 1991-1996.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
^{7}Be	0.6	0.7	0.8	1.0	1.4	1.4	1.2	1.2	1.1	1.0	0.8	0.6
^{137}Cs	1.3	1.6	1.3	0.9	1.1	1.4	0.8	0.5	0.7	0.7	0.7	1.3

Table 2. The average quarterly seasonal indices, 1991-1996.

Season	Winter	Spring	Summer	Fall
^{7}Be	0.63	1.07	1.27	0.97
^{137}Cs	1.40	1.10	0.90	0.76
Precipitation	0.60	0.90	1.24	0.82

The spring to fall periods of increased ^{7}Be concentrations and the winter periods of decreased ^{7}Be concentrations are clearly recognized. The lowest concentrations were obtained in December and the highest in May-June, with a steeper increase during late spring and a slower decrease in late fall and winter.

The highest concentrations of ^{137}Cs were obtained in winter periods when agricultural activities in the surrounding fields induced local surface dust resuspension effects, while maxima in June-July could be attributed to air exchange weather conditions. Seasonal variations of ^{137}Cs are less uniform and more dependent on unusual meteorological conditions in the period (high precipitation in the summers of 1991, 1992, and 1994).

The seasonal variations of ^{7}Be and ^{137}Cs air concentrations could also be analyzed by seasonal indices, defined as ratios of mean monthly concentrations and the overall average concentration in the period. The seasonal indices for ^{7}Be and ^{137}Cs are presented in Table 1, while quarterly (winter: December, January, and February; spring: March, April, and May; fall: September, October, and November; summer: June, July, and August) seasonal indices for ^{7}Be, ^{137}Cs, and the amount of precipitation are presented in Table 2.

Analysis of the seasonal indices data confirmed the seasonal variation pattern for both radionuclides and the maximum/minimum concentrations.

No correlation between the ^{137}Cs concentrations and the amount of precipitation was obtained. The washout effect of rainfalls seems to be more closely related to the variations in ^{7}Be ground air concentrations, but the correlation is still poor (coefficient of correlation $r<0.5$). The migration from stratosphere to troposphere is likely to play a crucial role. Also, there was no correlation between the ^{7}Be and ^{137}Cs concentrations.

The max/min ratios (ratios of maximum to minimum concentrations in the year) for ^{7}Be and ^{137}Cs in summers and winters are presented in Fig. 7 (the missing data were normalized to the overall average concentrations). A relatively stable pattern of ^{7}Be behaviour is observed, confirming the relatively constant rate of ^{7}Be production. The trend of the ^{137}Cs summer max/min ratios points to the constant slow decrease of Chernobyl-originated cesium, while the variations in ^{137}Cs winter max/min ratios confirm that this effect can be attributed to local climate phenomena.

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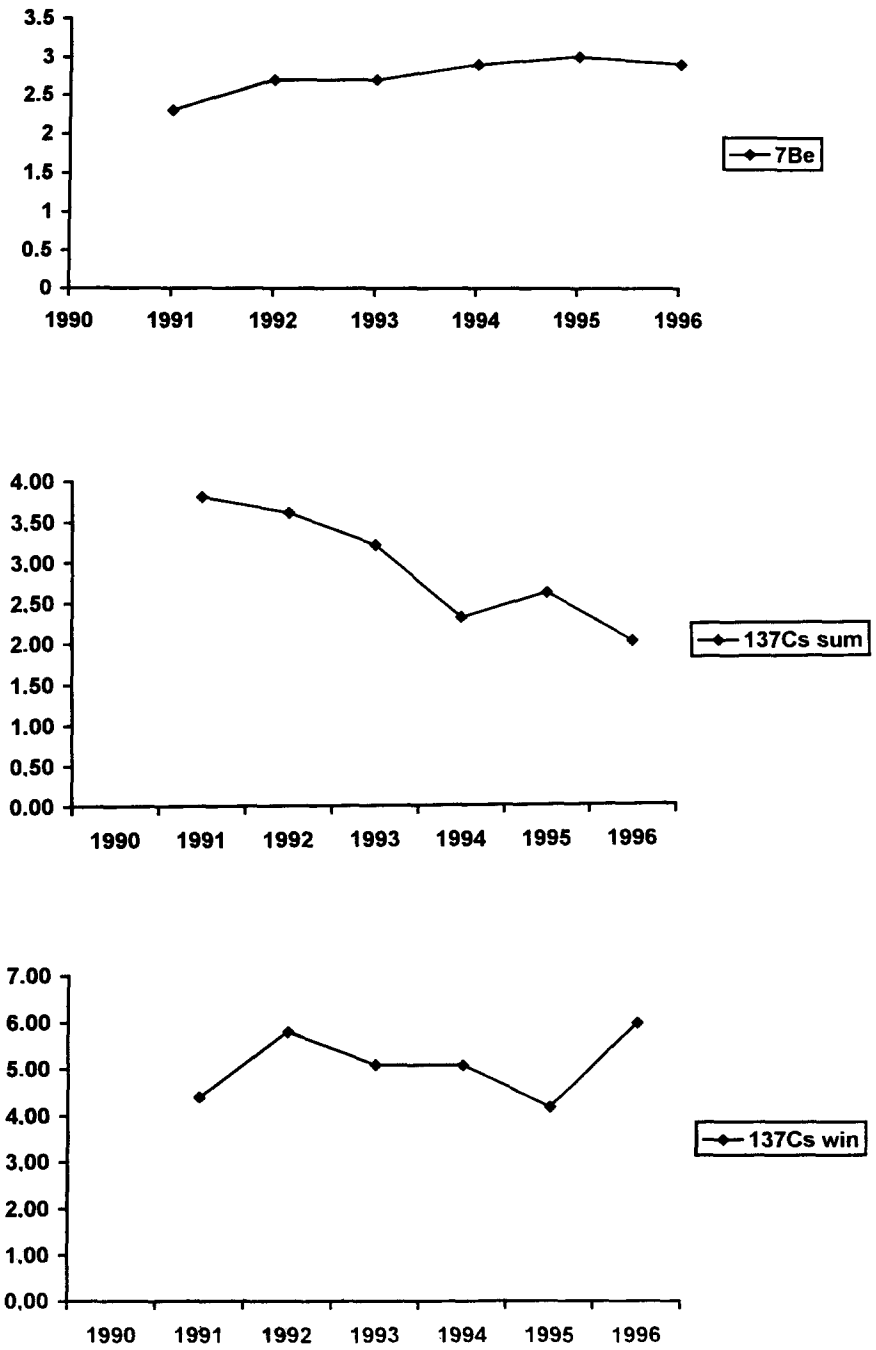

Fig. 7. Max/min ratios for ^{7}Be and ^{137}Cs in the period 1991-1996.

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