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RADIOACTIVE MATTER IN THE ZAGREB AIR FROM 1961 TO 1996

Radioaktivne tvari u zraku u Zagrebu od 1961. do 1996. godine.

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Abstract: This paper presents the results of long–term investigations of radioactive matter gathered from the air in the city of Zagreb. The total beta–activity in the air has been measured ever since 1961. A gamma–spectrometric air analysis has been carried out continuously since 1983, and the determination of ^{90}Sr since 1987. The data presented data indicate that the radioactive matter in the Zagreb air, as well as in the whole of Croatia, from 1961 to 1996, has been manifesting a constant exponential decrease in quantity. The only exception was naturally produced ^{7}Be, with almost unchanged specific activity concentrations throughout the measurement period. The main sources of radioactive matter in the air are still the nuclear explosions conducted in the atmosphere in the 1960s. The Chernobyl nuclear accident caused a significant increase in air radioactivity only in 1986. However, in the few subsequent years it decreased to pre–Chernobyl values. Despite the constant presence of radioactive matter in the Zagreb air, in the last thirty–six years, activity values have never exceeded the legal limit.

Key words: radioactivity, air, derived activity concentrations, Chernobyl

Sažetak: U radu su opisani rezultati dugogodišnjih istraživanja radioaktivnih tvari u zraku sakupljanom u Zagrebu. Ukupna beta–aktivnost mjeri se od 1961. godine. Gamaspektrometrijska analiza kontinuirano se provodi od 1983. a određivanje ^{90}Sr od 1987. godine. Prikazani podaci ukazuju na eksponencijalni pad radioaktivnih tvari u zagrebačkom zraku, kao i u cijeloj Hrvatskoj u periodu od 1961. do 1996. godine. Jedini je izuzetak prirodni ^{7}Be, čije su specifične aktivnosti u cijelom promatranom periodu gotovo nepromijenjene. Najvažniji izvor radioaktivnih tvari u zraku još su uvijek atmosferske nuklearne eksplozije iz početka 1960–tih godina. Nuklearna nesreća u Černobilju prouzročila je znatniji porast radioaktivnosti u zraku samo 1986. godine. No već sljedećih godina opet je opala na pretčornobiljske vrijednosti. Usprkos stalnom prisustvu radioaktivnih tvari u zagrebačkom zraku, aktivnosti u proteklih trideset šest godina nikada nisu premašile zakonom dopuštene vrijednosti.

Ključne riječi: radioaktivnost, zrak, izvedene koncentracije aktivnosti, Černobilj

1. INTRODUCTION

Long range atmospheric transport processes can cause a widespread global distribution of radioactive matter although it sometimes, like in Chernobyl, originates at a single point. Such radioactivity eventually reaches men, either directly or indirectly through the food chain. In order to study the fate and distribution of radionuclides introduced in the atmosphere, as well as to assess their impact on humans, it is necessary to measure the radioactivity in the air (Cesar et al. 1992, Cesar and Senčar 1996).

In the 1960s measurements of the total beta activity in the air were the mostly utilized met-

hod worldwide for the continuous monitoring of radioactive contamination in the air. However, in the late 1970s, due to scientific advancements in the field of electronics, gammaspectrometric measurements became the basic method for the qualitative and quantitative determination of radionuclides in various samples, including air. In addition, gammaspectrometry has numerous advantages compared to the total beta–activity measurements. The latter method cannot discriminate between the contribution of particular radionuclides and, to get more information, a time–consuming and tedious radiochemical analysis has to be performed. On the contrary, gammaspectrometrical measurement simultaneously gives information about the presence of different radionuclides. As radionuclides are mostly gamma emitters, only few important ones are left that cannot be determined gammaspectrometrically. These are ^{89}Sr, ^{90}Sr, ^{239}Pu, ^{240}Pu, ^{14}C and ^{3}H. Fortunately, these radionuclides are not very volatile (except ^{3}H). Therefore, they are not expected to be released in the environment in major quantities even in the case of severe nuclear accidents.

The fission radionuclides (thirty–five) routinely gammaspectrometrically measured in the air as a part of the monitoring programme of environmental radioactive contamination in Croatia are: ^{24}Na, ^{51}Cr, ^{54}Mn, ^{56}Mn, ^{57}Co, ^{58}Co, ^{60}Co, ^{59}Fe, ^{65}Zn, ^{88}Rb, ^{95}Nb, ^{95}Zr, ^{99}Mo, ^{99m}Tc, ^{103}Ru, ^{106}Ru, ^{110m}Ag, ^{124}Sb, ^{125}Sb, ^{129}I, ^{131}I, ^{132}I, ^{133}I, ^{134}I, ^{135}I, ^{132}Te, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{138}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce and ^{239}Np. In addition, eleven natural radionuclides are also measured: ^{7}Be, ^{40}K, ^{208}Tl, ^{212}Bi, ^{214}Bi, ^{214}Pb, ^{226}Ra, ^{228}Ac, ^{234m}Pa, ^{234}Th and ^{235}U.

Investigations of the distribution and fate of natural, weapon–produced and reactor–released radionuclides in the Zagreb air have been conducted by the Radiation Protection Unit of this Institute, as part of an extended monitoring programme. The results have been published yearly (Popović 1962–1978, Bauman et al. 1979–1992, Kovač et al.1993–1997). The total beta activity in the air in Croatia has been measured ever since 1961, while gammaspectrometric measurements started in 1981. Investigations of ^{90}Sr involving radiochemical methods started in 1987, a year after the Chernobyl nuclear accident. In addition to Zagreb, the radioactive contamination of air has been monitored over shorter or longer periods of time at some other locations in Croatia: Zadar (1964–1996), Pušća (1994–1996), Medvednica — Sljeme (1962–1964), Virovitica (1969–1990), Bistra (1981–1993), Stružec (1985–1987), Gospić (1986–1990) and Plomin (1987–1990).

Among anthropotropic (i.e. man–made) radioactive nuclides fission products, ^{137}Cs and ^{90}Sr have been regarded as fission products of great potential hazard to living beings due to a unique combination of their relatively long half–lifes and their chemical and metabolic properties resembling those of potassium and sodium, respectively.

^{137}Cs is a beta emitter with a half–life of 30 years. Its radioactive daughter, ^{137m}Ba, is a gamma emitter with a half–life of 3 minutes. Caesium is an alkaline metal like potassium and it resembles potassium metabolically. It belongs to the category of highly radiotoxic nuclides (group II) (ICRP 1983, S.l. SFRJ 1986, S.l. SFRJ 1984, N.N. 1991).

^{90}Sr has a radioactive half–life of 29.12 years and decays by beta emission. Its daughter ^{90}Y is also radioactive, with a half–life of 64 hours, and decays by beta emission to the stable isotope ^{90}Zr. Strontium is an alkaline earth element and is similar to calcium, barium and radium. Therefore, it follows calcium through the food chains from environment to man. Like calcium, strontium is retained in the body, mainly in the bones. Therefore, ^{90}Sr belongs to the category of highly radiotoxic nuclides (group I) (ICRP 1983, S.l. SFRJ 1986, S.l. SFRJ 1984, N.N. 1991).

^{7}Be is a gamma emitting radionuclide with a half–life of 53.28 days. It belongs to the third group of moderately radiotoxic nuclides (group III) (ICRP 1983, S.l. SFRJ 1986, S.l. SFRJ 1984, N.N. 1991).

2. MATERIAL AND METHODS

The total beta–activity in the air in Croatia has been measured ever since 1961. Air sampling, ranging from 100 to 200 m^3 daily, is carried out 1 m above the ground. Air is continuously pumped through the Schneider & Poelman filter paper (blue). The activity is measured after 120 h (in order to allow the decay of short–lived beta radionuclides) by a low level anti–coincidence, shielded beta counter. The mean values of daily total beta–activities were calculated for the respective years.

Gamma–spectrometric air analysis has been carried out continuously since 1983. Air is pumped through an air glass–fiber filter device placed 1 m above ground, the monthly volume ranging from 10,000–30,000 m^3. A gamma–ray spectrometry system based on a Ge(Li) detector (relative efficacy 15.4% with a FWHM resolution of 1.87 keV at 1.33 MeV) coupled to a computerized data acquisition system (a 4096–channel pulse height analyzer and a personal computer) was used to determine radioactivity levels in the samples from their gamma–ray spectra. The detector is shielded with 10 cm thick lead lined with 2 mm of cadmium and 2 mm of copper. Samples were measured in cylindrical plastic containers of appropriate volume, placed directly on the detector. The same samples were afterwards used for radiochemical separation (Harley 1970) of ^{90}Sr. The radioactivity of ^{90}Sr was determined by beta–counting its decay product (^{90}Y), in a low–background anti– coincidence, Geiger–Müller counter.

The mean values of monthly ^{90}Sr, ^{137}Cs and ^{7}Be specific activities were calculated for the respective years.

The counting time of all samples depended on sample activity concentration, but was never less than 60,000 s. Efficiency calibrations were carried out using sources provided by the International Atomic Energy Agency (IAEA) and the World Health Organization (WHO).

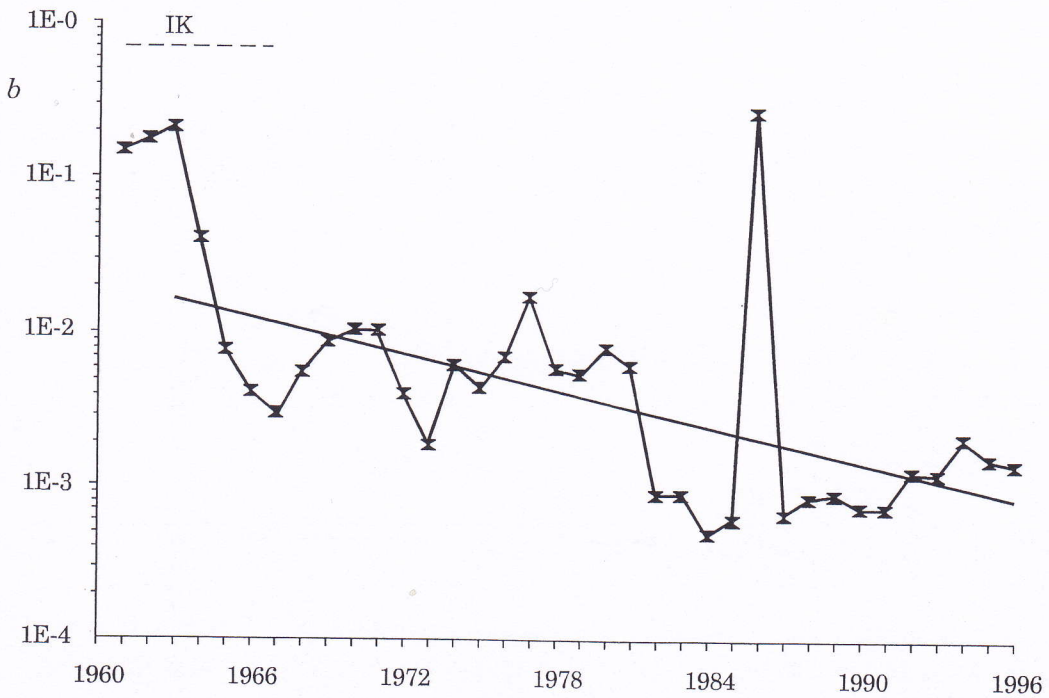

Figure 1. Total beta–activity (*b*) expressed in Bqm^{-3} in the Zagreb air from 1961 to 1996 with the pertaining regression curve

Slika 1. Ukupna beta–aktivnost (*b*) u Bqm^{-3} u zraku grada Zagreba od 1961. do 1996. godine i pripadajuća regresijska krivulja — pravac

3. RESULTS AND DISCUSSION

The moratorium on atmospheric testing of nuclear weapons in the early 1960s a resulted in constant decrease of radioactive matter in the air, as seen from Figure 1, showing long–term data of the total beta–activity in the Zagreb air. Although some other mathematical models can lead to a better fit of experimental data, the best physically acceptable fit for the experimental data is the exponential function:

$$A(x) = A(0)e^{-kx} \qquad (1)$$

where $A(x)$ and $A(0)$ are the activity concentrations at times x and zero, respectively, and k is the constant. The reciprocal value of the constant k is the mean residence time T_m of the observed radionucide in the air, i.e.:

$$T_m = \frac{1}{k} \qquad (2)$$

The transient increases in Figure 1 can be explained by the smaller–scale tests performed by China and France in spite of the nuclear moratorium, as well as by a variety of environmental physical factors that naturally fluctuate. For the total beta activity in the air, the fit of experimental data to a function (1) was obtained by regression analysis, leading to the equation:

$$b(x) = 0.018e^{-0.090x} \quad \left(\mathrm{Bqm}^{-3}\right) \qquad (3)$$

with $P(t)<0.001$. The time is in years, the starting year $x=1$ being 1963. The 1986 activity peak is caused by the presence of Chernobyl radionuclides like ^{137}Cs and ^{131}I, as confirmed by gammaspectrometric measurements. However, a year later, the total beta–radioactivity levels decreased to pre–Chernobyl values. Therefore, for the overall observed period, this peak did not influence the significance of the regression curve as indicated by the Fischer t–test.

Figure 2 shows the specific ^{137}Cs activity concentrations in the Zagreb air from 1983 to 1996. The 1986. activity peak was caused by ^{137}Cs originated in the Chernobyl accident. The data for ^{137}Cs specific activity concentrations are fitted

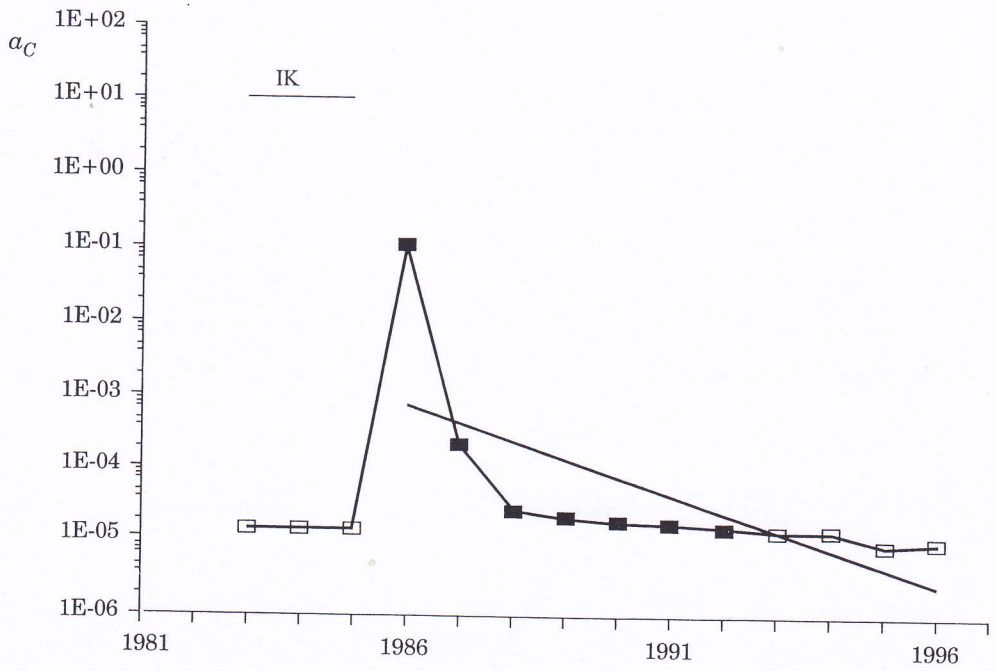

Figure 2. ^{137}Cs specific activity concentrations (a_C) expressed in Bqm^{-3} in the Zagreb air from 1983 to 1996 with the pertaining regression curve

Slika 2. Specifične aktivnosti ^{137}Cs (a_C) u Bqm^{-3} u zraku grada Zagreba od 1983. do 1996. godine i pripadajuća regresijska krivulja — pravac

to the exponential function:

$$a_c(x) = 0.014e^{-0.567x} \quad \left(\text{Bqm}^{-3}\right) \qquad (4)$$

with $P(t)<0.02$. The time is in years, the starting year $x=1$ being 1986.

From equations (2) and (4) the mean residence time of ^{137}Cs for the post–Chernobyl period, characterized by a rapid decrease of ^{137}Cs in the air, is 1.8 years. In comparison, the mean residence time of ^{137}Cs in fallout for the period May 1986 – December 1990 is 0.71 years (Franić, 1992).

Figure 3 shows specific ^{90}Sr activity in Bqm^{-3} in the Zagreb air from 1987 to 1996. The best fit for the ^{90}Sr specific activity concentrations is the exponential function

$$a_s(x) = 0.00003e^{-0.340x} \quad \left(\text{Bqm}^{-3}\right) \qquad (5)$$

with $P(t)<0.01$. The time is in years, the starting year $x=1$ being 1987.

From equations (2) and (5) the mean residence time of ^{90}Sr in the air is 2.9 years. In comparison, the mean residence time of ^{90}Sr in fallout, for the period 1962–1993 is 5.6 years. However, the ^{90}Sr mean residence time for the period 1962–1967, which followed the most intensive atmospheric nuclear weapon tests, is only 1.27 years (Franić, 1994). Namely, like the post–Chernobyl period for ^{137}Cs, the 1962 –1967 period is characterized by a rapid decrease of ^{90}Sr activity concentrations in the air.

The nuclear accident at Chernobyl (26 April 1986) did not cause a significant increase in ^{90}Sr

Table 1. Total beta–activity expressed through the percentage of a derived concentration of an unknown mixture of radionuclides and specific ^{7}Be, ^{137}Cs and ^{90}Sr activities expressed through the percentage of a corresponding derived concentration in the Zagreb air from 1961 to 1996

Tablica 1. Postoci ukupne beta–aktivnosti (b) u odnosu na izvedenu koncentraciju za nepoznatu smjesu radionuklida i specifičnih aktivnosti ^{7}Be, ^{137}Cs i ^{90}Sr u odnosu na odgovarajuće izvedene koncentracije u zraku grada Zagreba od 1961. do 1996. godine (%IK)

	b		b	^{7}Be	^{137}Cs	^{90}Sr
IK/Bqm^{-3}	0.7		0.7	2000	10	0.4
Year	%IK	Year	%IK			
1961	214	1979	0.793	—	—	—
1962	25.4	1980	1.14	—	—	—
1963	30	1981	0.886	—	—	—
1964	5.81	1982	0.129	—	—	—
1965	11.1	1983	0.129	0.0002	<1.5E–4	—
1966	0.581	1984	0.0714	0.0002	<1.5E–4	—
1967	0.423	1985	0.0886	0.0004	<1.5E–4	—
1968	0.793	1986	38.6	0.0004	1.1	—
1969	1.27	1987	0.0957	0.0006	0.0022	0.008
1970	1.49	1988	0.12	0.0002	0.0003	0.0023
1971	1.49	1989	0.127	0.0003	0.0002	0.0022
1972	0.581	1990	0.106	0.0002	0.0002	0.0045
1973	0.264	1991	0.104	0.0002	0.0002	0.0018
1974	0.896	1992	0.179	0.0003	0.0002	0
1975	0.634	1993	0.171	0.0003	<1.4E–4	0
1976	1	1994	0.29	0.0003	<1.4E–4	0
1977	2.49	1995	0.213	0.0003	<9.2E–5	0
1978	0.846	1996	0.197	0.0003	<1.0E–4	0

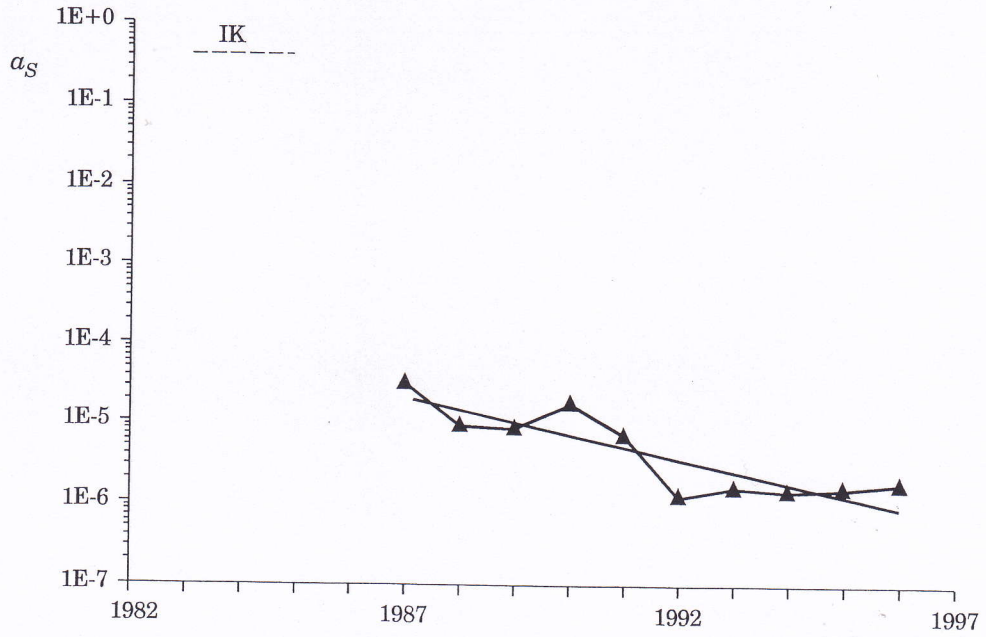

Figure 3. ^{90}Sr specific activity concentrations (a_S) expressed in Bqm^{-3} in the Zagreb air from 1987 to 1996 with the pertaining regression curve

Slika 3. Specifične aktivnosti ^{90}Sr (a_S) u Bqm^{-3} u zraku grada Zagreba od 1987. do 1996. godine i pripadajuća regresijska krivulja — pravac

activity in the environmental samples in Croatia (including air), as opposed to ^{137}Cs. Unlike the debris from the atmospheric testing of nuclear weapons, the radionuclides that originated from the Chernobyl damaged reactor were not released directly into the upper atmosphere. As the result of the release mechanism (continuing releases over a 10–day period, steam explosions and fire of the graphite moderator) and the prevailing meteorological conditions at the time, the less volatile components of the Chernobyl debris (e.g. ^{90}Sr) were deposited closer to the accident location than the more volatile constituents, e.g. radiocaesium (Whitehead et al. 1988). Thus, ^{90}Sr was not subjected to global dispersion processes, being deposited to the Earth's surface within a period of a few days to a few weeks after the accident. Generally, changing meteorological conditions with wind from different directions at various altitudes and prolonged releases resulted in a very complex dispersion pattern over Europe. Consequently, major parts of Croatia were initially unaffected by the plumes of contaminated air (UNSCEAR 1988, UNEP 1991).

The specific activity concentrations of ^{7}Be in the Zagreb surface air during the study period 1983–1996 are shown in Figure 4.

The lines present the mean value and the corresponding standard deviations. For the observed 1982–1996 period, no trends in ^{7}Be specific activity concentrations can be observed. The mean value ($\bar{a}_B$) for the 1982–1996 period is $(6 \pm 2) \times 10^{-3}$ Bqm^{-3} with a 33% standard error (g_s). Similar values have been observed elsewhere (Paatero et al. 1996). A relatively high standard error indicates pronounced fluctuations in ^{7}Be specific activity concentrations. These fluctuations can be explained by the mechanism of production and transport of ^{7}Be to the Earth surface. ^{7}Be, being a „cosmogenic radionuclide" is produced in the upper troposphere and stratosphere in nuclear reactions induced by cosmic rays in atmospheric oxygen and nitrogen atoms (Lujanas 1979). About 75% of ^{7}Be is produced in the stratosphere and 25% in the upper troposphere (Johnson and Viezee 1981). The descending movement of air during high–pressure situations subsequently brings ^{7}Be downwards. Therefore, ^{7}Be activity concen-

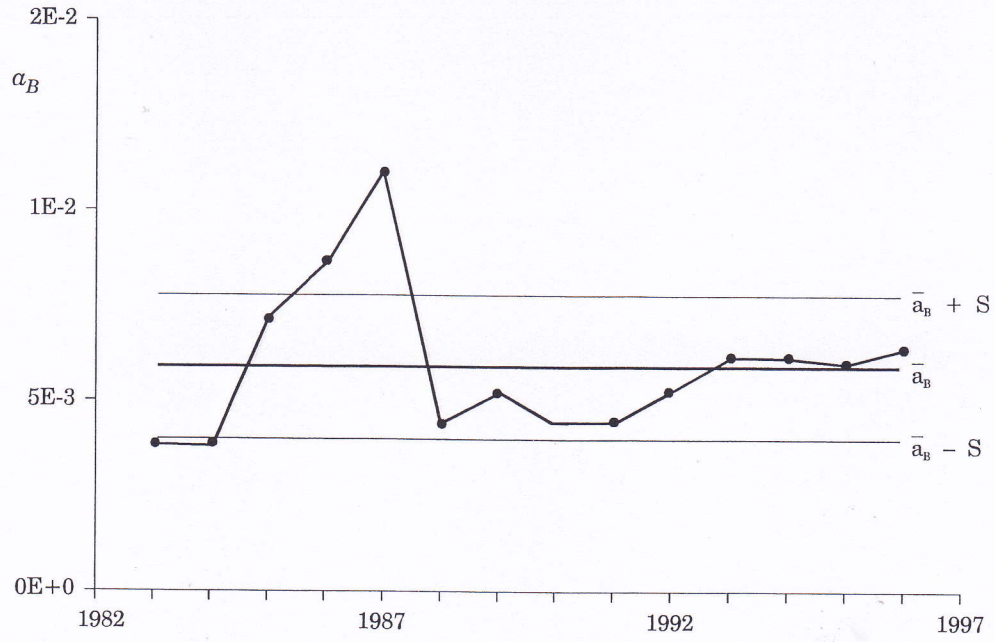

Figure 4. ^{7}Be specific activity concentrations (a_B) expressed in Bqm^{-3} in the Zagreb air with the pertaining average and standard deviation

Slika 4. Specifične koncentracije aktivnosti ^{7}Be (a_B) u Bqm^{-3} u zraku grada Zagreba i pripadajuća prosječna vrijednost sa standardnom devijacijom

trations in the near–surface air are in a very complex way dependant upon meteorological conditions.

To conveniently present the impact of the radioactive matter in the air on humans, the data of the total beta–activity measured and the specific activities of ^{7}Be, ^{137}Cs and ^{90}Sr are shown in Table 1 as percentages of derived air concentrations (%IK), i.e. as the limit values determined by Croatian law. (S.l. SFRJ 1987, S.l. SFRJ 1984, N.N. 1991). It can readily be seen that, for the whole observed period, in the surface air in Zagreb the concentrations of radioactive matter never exceeded allowed values.

4. CONCLUSIONS

On the basis of presented data, it can generally be concluded that for the whole 1961–1996 period the concentrations of radioactive matter in the Zagreb air exponentially decreased, the only exception being activity concentrations of naturally produced ^{7}Be. The nuclear accident in Chernobyl caused an activity peak in 1986. However, in the subsequent years its influence on the quantities of radioactive matter in the air was negligible. Therefore, the main source of radioactive matter in the air are still the atmospheric nuclear explosions performed in the 1960s.

Continuous monitoring during the observed period showed that the activity concentrations of the radioactive matter in the Zagreb air never exceeded allowed values. However, in order to protect the general public, monitoring is necessary, also as part of a general emergency preparedness in case of a nuclear accident.

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