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LONG-TERM INVESTIGATIONS OF RADIOACTIVE MATTER IN THE AIR IN THE CITY OF ZAGREB

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ABSTRACT

The activity concentrations of radioactive matter in the Zagreb have been manifesting the constant exponential decrease ever since early 1960s. The Chernobyl nuclear accident caused a major increase in Zagreb air radioactivity only in 1986, which over next few years quickly decreased to pre-Chernobyl values. However, due to changing meteorological conditions at that time, resulting in very complex dispersion pattern of Chernobyl debris over Europe, major parts of Croatia have been initially unaffected by the plumes of contaminated air.

The mean residence time of ^{90}Sr and ^{137}Cs in fallout are estimated for respective pre-Chernobyl and post-Chernobyl periods.

Despite of constant presence of radioactive matter in the Zagreb air, in the observed period activity concentration values never exceeded the legal limits. Consequently, the doses incurred by ^{90}Sr and ^{137}Cs by inhaling the polluted air after the Chernobyl accident are very small.

Key words: radioactivity, ^{90}Sr, ^{137}Cs, Chernobyl accident, dose

1. INTRODUCTION

Nuclear tests conducted in the atmosphere and releases of radioactive material from nuclear facilities are the main causes of the man-made radioactive contamination of human environment. Once released to the atmosphere, long-range atmospheric transport processes can cause a widespread distribution of such radioactive matter, although it may, like in the case of Chernobyl accident, originate in a single point.

The resulting fallout, consisting of short and long-lived radionuclides, eventually affects humans, either directly or indirectly by entering the food chain through the plants and animals. In both cases it causes a health hazard to the population either through the direct irradiation or consumption of contaminated foodstuffs.

Among man - made radioactive nuclides, those of the strontium and caesium, particularly ^{90}Sr and ^{137}Cs, are regarded as a great potential hazard to living beings. Namely, these fission products have unique combinations of relatively long half-lives (29.1 and 30.14 years respectively) and chemical and metabolic properties resembling those of the potassium and calcium respectively.

Investigations of the distribution and fate of natural, nuclear weapons produced and reactor released radionuclides in the Zagreb air have been conducted as a part of an extended and still ongoing monitoring programme of radioactive contamination of human environment in Croatia. The gross beta activity in air has been measured ever since 1961, while systematic gammaspectrometric measurements started in 1983. Before that time and starting in 1965, ^{137}Cs activity concentrations were measured by NaI detector. Investigations of ^{90}Sr, involving radiochemical methods, started in 1987, i.e., one year after the Chernobyl accident. The measurements of external ambient gamma dose rates have been performed since 1985 ((Popovic 1966-1978; Bauman et al. 1979 – 1992; Kovac et al. 1993 – 1998; Marovic et al.1999 - 2005).

2. MATERIAL AND METHODS

Air samples have been collected daily 1 m above the ground by a high volume air sampler that pumps air through glass-fiber filter. Total volume of air passing through the filters is 40,000 to 50,000 m^3.

Fallout samples were collected monthly in the city of Zagreb at the location of the Institute for Medical Research and Occupational Health (45° 50' 7.3" N, 15° 58' 58.7" E). The funnels that were used for fallout collection had a 1 m^2 catchment area. Precipitation quantity was measured by Hellman pluviometer.

A gamma-ray spectrometry system based on a low-level ORTEC Ge(Li) detector (FWHM 1.87 keV at 1.33 MeV ^{60}Co and relative efficacy of 15.4% at 1.33 MeV) coupled to a computerized data acquisition system was used to determine radiocaesium and beryllium-7 levels in the samples from their gamma-ray spectra. Samples were measured in cylindrical plastic containers of appropriate volume, which were placed directly on the detector. Counting times depended on sample activities, ranging from 10,000 to 250,000 seconds, typically being 80,000 s.

The activity concentrations of ^{90}Sr were determined after the radiochemical treatment of samples, by beta counting its decay product, (^{90}Y), in a low-background, anti-coincidence shielded Geiger-Müller counter.

Quality assurance and intercalibration measurements were performed through participation in an International Atomic Energy Agency (IAEA) and World Health Organization (WHO) international intercalibration programmes.

3. RESULTS AND DISCUSSION

^{137}Cs activity concentrations in air and fallout

The radioactive fallout resulting from large-scale nuclear weapon tests in the atmosphere conducted in the 1960s, followed by similar, but smaller scale tests by the Chinese and French in the 1970s and afterwards, was the dominant route for the introduction of artificial radionuclides in the environment until the nuclear accident at Chernobyl, in former USSR, on 26 April 1986. Therefore, activity of most

environmental samples could be expected to be in correlation with fallout activity (i.e. surface deposit in Bqm^{-2}).

Severe radioactive fallout from highly radioactive air plumes that originated from the damaged Chernobyl nuclear reactor was spread and transported all over Europe. Fortunately, due to the prevailing meteorological conditions at the time after the accident that influenced the formation and spreading direction of Chernobyl plumes, Croatia was only on the North-Western region partially affected by the edge of one of the plumes (UNSCEAR, 1988), as indicated on Figure 1.

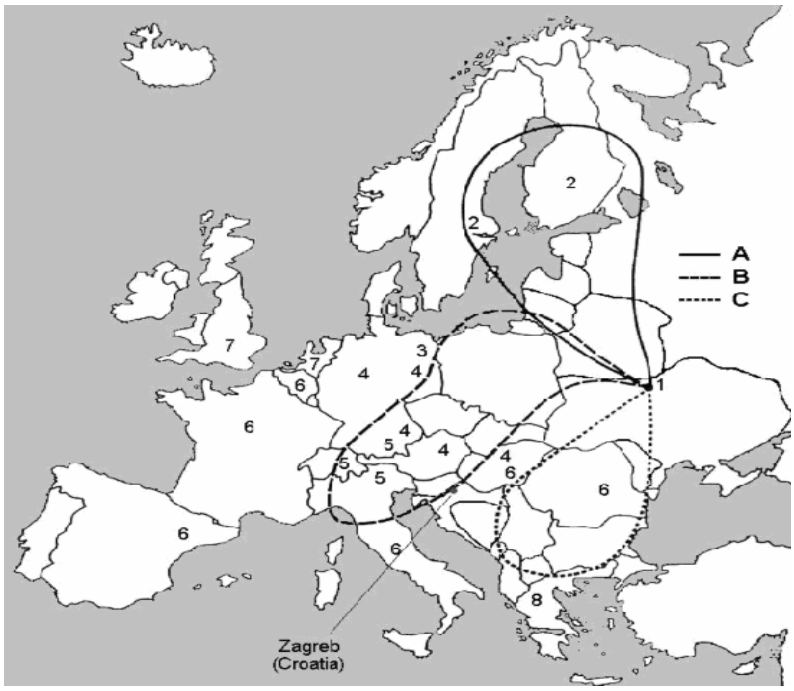

Fig. 1. Spreading of radioactive plumes over the Europe after the Chernobyl nuclear accident. Numbers 1-8 represent plume arrival times at respective areas: 1 = April 26, 2 = April 27, 3 = April 28, 4 = April 29, 5 = April 30, 6 = May 1, 7 = May 2 and 8 = May 3. The figure has been adopted from UNSCEAR report for 1988.

The highest ^{137}Cs activity concentrations in the observed period were recorded in May 1986, being 6200 Bqm^{-2} for the surface deposit by fallout and 0.85 Bqm^{-3} in air. Such high values could be attributed to the second of the three radioactive plumes originating from the Chernobyl reactor, which edge passed over Northwest Croatia (UNSCEAR 1988). The radioactive material introduced to the atmosphere by Chernobyl accident was by global dispersion processes distributed throughout the troposphere, causing the increased radiocaesium activity concentrations in the environment in years to come. However, ^{137}Cs these showed a significant exponential decrease over time because of natural removal as well as radioactive decay. Also, no new releases of ^{137}Cs occurred after the Chernobyl reactor accident either from nuclear facilities or nuclear weapons testing.

In 2004, average ^{137}Cs activity concentration in air was 0.02 Bqm^{-3}, while total ^{137}Cs surface deposit by fallout was 2.07 Bqm^{-2}. Activity concentrations of ^{137}Cs in air and fallout are shown on figure 2.

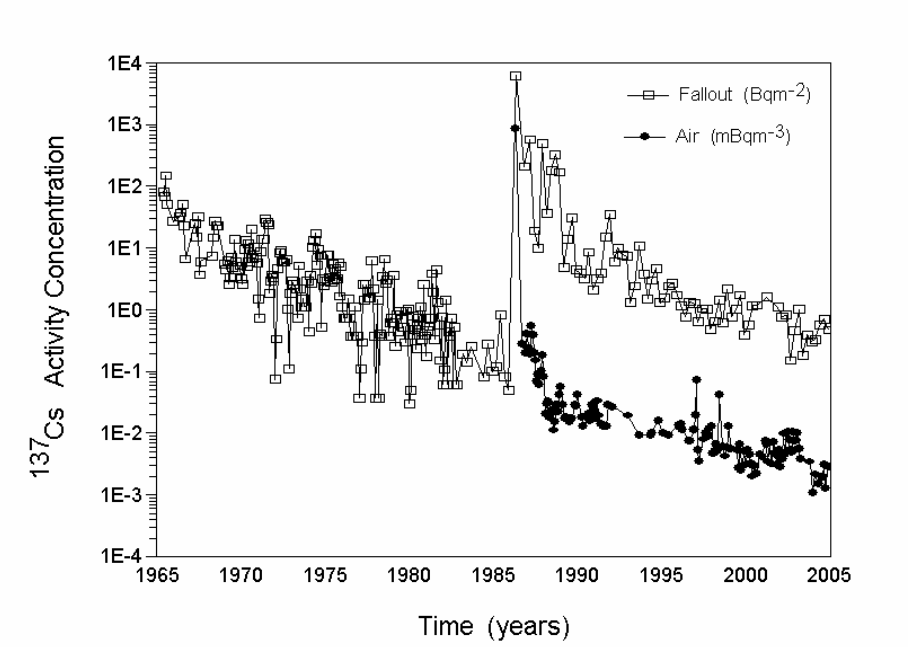

Fig 2. ^{137}Cs activity concentrations in air and fallout

The ^{137}Cs activity concentration in surface deposit in May 1965 was 81.4 Bqm^{-2}. The minimal value for the pre-Chernobyl period was recorded in December 1985, being 0.05 Bqm^{-2}, while the maximal value was 150.2 Bqm^{-2}, which was recorded in July 1965. The lower limit of total amount of ^{137}Cs surface deposit delivered by fallout for the period January 1965 – April 1986 was about 1300 Bqm^{-2}. This value is being regarded as a lower limit since some data are missing, which is especially true for the 1960s, when ^{137}Cs activity concentrations were quite large and it was no possible to include those data into the overall sum. For post-Chernobyl period, i.e. from May 1986 to December 2004, the lower limit of total amount of ^{137}Cs in surface deposit is 8450.5 Bqm^{-2}. However, if year 1986 is excluded, the total amount of ^{137}Cs in surface deposit is 2040.5 Bqm^{-2}.

^{7}Be activity concentrations in air

^{7}Be ($t_{1/2}$ = 53.3 days is naturally occurring radionuclide of cosmogenic origin, formed by spallation processes of light atmospheric nuclei such as nitrogen and oxygen when they capture protons or even neutrons from the primary component of cosmic rays. Once formed, ^{7}Be is rapidly attached to the particles in the atmosphere. Naturally produced ^{7}Be, with almost unchanged activity concentrations throughout the entire observed period, is a good quality check of the methods used.

In addition, this radionuclide has been recognized as a powerful tool in the description of environmental processes precipitation scavenging (i.e. washout), atmospheric particle deposition as well as deposition patterns of airborne contaminants. High ^{7}Be concentrations in air are related to low solar activity like in

1987, while low concentrations are related to high solar activities. The downward trend in the ^{7}Be air concentrations in 2004 can therefore be attributed to an increase in solar activity.

The mean ^{7}Be activity concentration in the air for the 1987 – 2004 period is (5.4 ± 2.8) $\times 10^{-3}$ Bqm^{-3}. ^{7}Be activity concentrations in air are shown on figure 3.

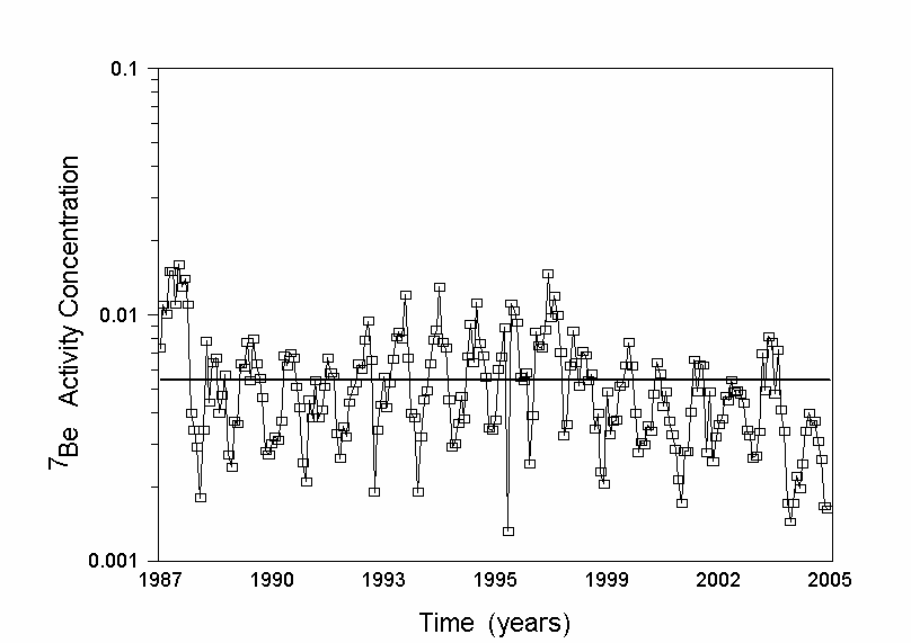

Fig 3. ^{7}Be activity concentrations in air

These values are very similar as observed elsewhere (Papastefanou et. al., 1995).

^{90}Sr activity concentrations in air and fallout

The minimal value of ^{90}Sr activity concentration in surface deposit delivered by fallout for the pre-Chernobyl period was recorded in October 1990, being 0.02 Bqm^{-2}, while the highest value was 247.9 Bqm^{-2}, which was recorded in June 1964. The total amount of ^{90}Sr surface deposit for the period January 1962 – April 1986 was 3782.8 Bqm^{-2} and 439.4 Bqm^{-2} for the post-Chernobyl period, i.e. from May 1986 to December 2004.

It should be noted that ^{90}Sr fallout peak, attributed to Chernobyl accident in Croatia decreased very rapidly, as in August 1986 was recorded only 1.81 Bqm^{-2} and 0.13 Bqm^{-2} in November.

Regarding the ^{90}Sr activity concentrations if air, the highest value for the overall observed period was recorded in the first quarter of 1987, being 44.6 mBqm^{-3}. The minimal value of 0.15 μBqm^{-3} was recorded in 2004.

It should be noted that in late 1990s and afterwards, ^{90}Sr activity concentrations approached very low values, being essentially background variations. Therefore,

transient increases and decreases in activity concentrations can be partially explained by a variety of environmental physical factors that naturally fluctuate.

^{90}Sr activity concentrations in air and fallout are shown on figure 4.

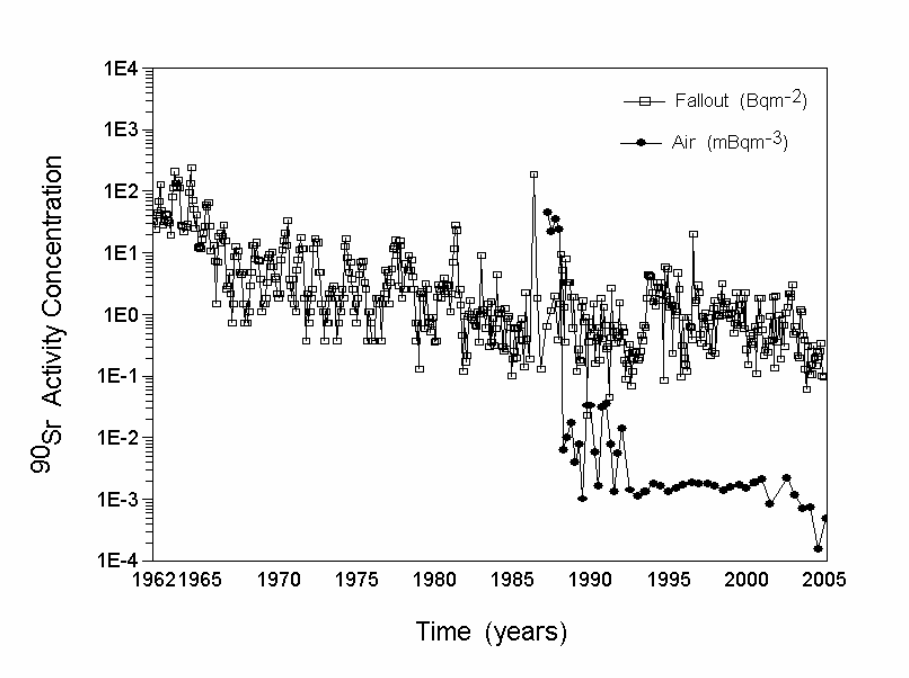

Fig 4. ^{90}Sr activity concentrations in air and fallout

Dosimetry of ^{90}Sr and ^{137}Cs

Data on activity concentrations of ^{90}Sr and ^{137}Cs in air allow for the estimate of the doses incurred by inhaling the polluted air. Dose conversion factors, i.e. effective dose per unit intake via inhalation for the member of public older than 17 years and moderate absorption, are 3.6×10^{-8} and 9.7×10^{-9} SvBq^{-1} respectively (IAEA, 1996). As the ratio of dose conversion factors for ^{90}Sr and ^{137}Cs is ≈3.7, it implies that inhaling ^{90}Sr contributes 3.7 times more to the dose, compared with inhaling the same activity concentration of ^{137}Cs.

The data on volume of breathed air have been taken from the International Commission on Radiological Protection (ICRP) publication 23 (1974). The ICRP reference man breathes 7.5 Lmin^{-1} during rest and 20 Lmin^{-1} during light activity (ICRP, 1974). As the daily activities include approximately 8 h resting and 16 h of light activities, the total daily volume of breathed air is 22,800 L.

Therefore, the annual effective dose in year 1987, incurred by inhaling ^{90}Sr from air is 1.2×10^{-5} Sv and 3.1×10^{-9} Sv in year 1988, decreasing to only 9.2×10^{-11} Sv in year 2004. Regarding ^{137}Cs, similar analysis yields to 1.7×10^{-4} Sv in 1986, 8.23×10^{-8} Sv in 1987, 1.1×10^{-8} in 1988, decreasing to only 7.4×10^{-10} Sv in 2004.
As the doses incurred by ^{90}Sr and ^{137}Cs by inhaling the polluted air after the Chernobyl accident are very small, it can be argued that air was not the critical pathway for human intake of ^{90}Sr and ^{137}Cs from the environment.

Mean residence time of ^{90}Sr and ^{137}Cs in air and fallout

Decrease of activity concentrations of radioactive matter either in air or fallout can be described by the exponential function:

$$C_A(t) = C_A(0)\, e^{-kt} \qquad (1)$$

where:

$C_A(t)$ is time-dependant activity concentration of observed radionuclide (^{137}Cs or ^{90}Sr) in fallout or air (Bqm^{-2} for fallout and Bqm^{-3} for air),

$C_A(0)$ initial activity concentration of observed radionuclide in fallout or air (Bqm^{-2} for fallout and Bqm^{-3} for air) and

$1/k=t_o$ observed mean residence time of considered radionuclide in fallout or air (years).

By fitting the experimental data to the curve (1) the observed mean residence time for ^{137}Cs in air, for the immediate post-Chernobyl period, i.e. January 1987 – December 1990 was estimated to be 1.0 years, while the observed mean residence time for ^{137}Cs in fallout was estimated to be 0.9 years. The year 1986 was excluded from the analysis due to direct influence of the plume. For comparison, the observed mean residence time of ^{137}Cs in fallout for the period January 1965 – April 1986 was 3.7 years (Franic 1992).

Regarding ^{90}Sr, the observed mean residence time in air for the period January 1962 – April 1986 was estimated to be 5.95 years (Franic 1994). For the immediate post-Chernobyl period, i.e. May 1986 – December 1990 was estimated to be 1.9 years.

^{137}Cs : ^{90}Sr activity ratio in fallout

In early 1960s radiochemical analysis of most environmental samples studied in the investigations of the consequences of nuclear fallout yielded ^{137}Cs : ^{90}Sr activity ratios generally ranging between values 1 and 3 (UNSCEAR 1964). Since both of these radionuclides have inert gaseous precursors in their fission chains, and generally similar chemical characteristics, substantial fractionation from the time of their creation in the nuclear explosion is considered to be unlikely. Therefore, in 1960s the major part of the variation in observed ^{137}Cs : ^{90}Sr activity ratios in environmental samples reflect errors in analyses.

For the pre-Chernobyl period, ^{137}Cs : ^{90}Sr activity ratio in fallout samples collected in Zagreb was 1.27 ± 1.38. Interestingly, ^{137}Cs : ^{90}Sr activity ratio in Adriatic sea water was a little bit higher and far more constant, being 1.52 ± 0.40 (Franic and Bauman, 1993).

The Chernobyl nuclear accident has altered this ratio by several orders of magnitude. In 1986 this ratio in Zagreb fallout was 32 in May and 162 in November, increasing ever since April 1987, when it started to decrease. In 1990, it again reached pre-Chernobyl values being 1.51.

4. CONCLUSIONS

The main sources of radioactive matter in the Zagreb air are still the atmospheric tests of nuclear weapons conducted in the 1960s. Activity concentrations for ^{137}Cs and ^{90}Sr in radioactive fallout are exponentially decreasing. ^{137}Cs activity concentrations in fallout are decreasing with different rates for the pre-Chernobyl and post-Chernobyl period, the observed mean residence times in fallout being 3.7 and 0.9 years for the respective periods. The observed mean residence time for ^{137}Cs in air for post-Chernobyl period is approximately 1 year.

The mean residence time of ^{90}Sr in fallout is estimated to be 5.95 years for 1962 – April 1986 period. After the Chernobyl accident, which caused major increase of ^{90}Sr activity concentration in fallout only in May 1986, its activity concentrations soon reached pre-Chernobyl values, nowadays being only background variations.

The doses incurred by ^{90}Sr and ^{137}Cs by inhaling the polluted air after the Chernobyl accident are very small, and it can be argued that air was not the critical pathway for human intake of ^{90}Sr and ^{137}Cs from the environment.

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