Fluctuation of radiocaesium concentrations in the near-surface atmospheric layer in Białystok in the period 1992–1999

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Abstract Air samples were collected at an early-warning sampling and monitoring radioactive contamination station ASS-500. The results of the measurements are presented for radioactive aerosols collected on filters each week between 1992 and 1999. The large variations in activity concentration were observed in annual cycles. The maximum values exceeded the mean annual values by a factor of 2.8 to 7.3. Over the period of eight years the mean values of activity in air was found to decrease from 5.3 μBq m^{-3} in 1992 to 2.2 μBq m^{-3} in 1999. A good correlation between the Cs-137 activity and dust content in the air indicates that in the observed period the main source of Cs-137 activity is the dust with soil and vegetation particles. The effect of radiation from inhalation is negligible (626 pSv).

Key words air contamination • Chernobyl accident • Cs-137

Introduction

Until 1986, the Poland's environment was marked by man-made radioactive pollution resulting mainly from nuclear weapon testing (radioactive fallout). Between 1945 and 1980, 423 nuclear explosions were carried out [16], and a fraction of man-made radionuclides released during these tests was transferred to the troposphere and stratosphere. It is estimated that 9.6×10^{15} Bq of Cs-137 was retained in the stratosphere [16]. Subsequently, the radionuclides were transferred to the troposphere and then to the soil surface [15].

Radioactivity released during the Chernobyl accident migrated mainly to the troposphere, only a small fraction went to the stratosphere [8]. As a result, the distribution of atmospheric (environmental) contamination after 1986 differed from that resulting from the nuclear weapons tests. At present, it is only Cs-137, due to its long half-life ($T_{1/2}$=30.2 years), that may constitute some radiation hazard to man, and its presence in the atmospheric air may cause radiation hazard to the population through inhalation.

Three main pathways for Cs-137 in the inhaled air may be distinguished:
- local re-suspension, that is withdrawal of radiocaesium deposited on the soil surface [12];
- contribution from the contamination of the stratosphere resulting from the Chernobyl accident, which may now migrate to the near-surface air layer [11], and
- long-distance transport of contamination from heavily contaminated areas in Russia, Belarus and Ukraine [3].

The contributions from all the three pathways to the local Cs-137 airborne concentrations are now being carefully examined [5], and are the subject of the present work.

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Received: 10 September 2001, Accepted: 18 February 2002

Methods

Air samples were collected at the ASS-500 station situated in the Biophysics Department of the Medical Academy in Białystok [13]. This radioactive contamination monitoring station, was designed and constructed by the Central Laboratory for Radiological Protection (CLRP) in Warsaw. This station forces both large amounts of atmospheric air (400–800 m^3/h) through a filter on which dust and aerosols are deposited, and measures and records changes in the global radioactive contamination deposited on the filter.

In the present work, results of the measurements are presented for the radioactive aerosols collected on filters in one-week cycles between 1992 and the end of 1999. As a filtering material a PCV fiber high-efficiency collection filter (Petrianov's FPP-15-1.7 [9]) was used. Before the main measurement the filter was pressed to form a disk (51 mm in diameter and from 4 to 8 mm thick – depending on the amount of dust collected on the filter). Spectrometric measurements were carried out in the Gamma Spectrometry Laboratory of the Biophysics Department. The analysis of the radionuclide content in the samples was made by gamma-ray spectrometry using a solid-state coaxial germanium detector (relative efficiency: 15%) manufactured by Canberra Company. The detector was placed in a 10 cm thick lead castle, which was internally lined with sheets of aluminium and cooper.

The lower detection limits were estimated according to the method used by Currie [2]. The calculated values of MDC for Cs-137 in the disk geometry (51 mm in diameter) ranged from 0.2 to 0.6 μBq m^{-3} depending on the amount of the air filtered.

Results and discussion

The weekly changes in Cs-137 concentrations in particular years are shown in Fig. 1. Table 1 provides mean annual Cs-137 levels for the region of Białystok in 1992–1999 based on the measurements carried out at the ASS-500 station. It is worth noting that large variation in concentrations occurred. Particularly high values were found in May 1993 (34.5 μBq m^{-3}) and in September 1992 (22.9 μBq m^{-3}). In other years the maximum values exceeded the mean annual values by a factor of 2.8 to 7.3.

The Cs-137 concentrations in samples of the filters were correlated with the amount of airborne dust and with the presence of Be-7, a radionuclide of cosmogenic origin. The results of these correlations are shown in Figs. 2 and 3.

In 1999, a good correlation (determined by Pearson's method) was found between Cs-137 and dust content ($r=0.677, p<0.001$) and a weak correlation between Cs-137 and Be-7 ($r=0.24, p<0.09$), indicating that the presence of radiocaesium in the near-surface atmospheric layer is mostly due to the dust, containing soil and vegetation particles rising from the soil surface.

The presence of airborne Cs-137 leads to an increased radiation hazard to the population from inhalation. The basic quantity used to determine the above hazard is referred to as the effective inhalation dose, which may be expressed as the product of the following factors:

(concentration in air) × (breathing rate) × × (breathing time) × (inhalation dose conversion coefficient depending on the class of inhalation dose) = = (annual effective dose equivalent).

For the annual effective dose equivalent from Cs-137 in air the following assumptions have been adopted for adults:
- Cs-137 concentrations from Table 1; it was assumed that the activity concentrations of Cs-137 in-door was the same as out-doors;
- the breathing rate of 20 m^3/day for the whole population [17];
- the breathing time of 365 days, and
- the effective dose conversion coefficient per inhalation unit of 3.9×10^{-8} $SvBq^{-1}$ [6].

The results are given in Table 1. The dose from radiocaesium absorbed by inhalation ranged from 1509 pSv in 1992 to 626 pSv in 1999.

The presence of Cs-137 in the near-surface soil layer, apart from entering the alimentary chain through feed and plants suitable for consumption, is the main factor that determines the radiation hazard from post-Chernobyl gamma radionuclide inhalation. The Cs-137 levels in the near-surface atmospheric layer, detected from the measurements carried out at the ASS-500 station in Białystok, tended to fall. Over the period of eight years of measurements the activity was found to decrease from the mean value of 5.3 μBq m^{-3} in 1992 to the mean value of 2.2 μBq m^{-3} in 1999.

The assessment Cs-137 pathways of presented in this paper was based on the correlation analysis between airborne Cs-137 and Be-7, as well as between Cs-137 and dust content in air. The mean concentrations of radioberyllium (Be-7) detected in the investigated filters ranged from 2444 μBq m^{-3} in 1992 to 2263 μBq m^{-3} in 1999, the mean value over the years 1992–1999 being 2507 μBq m^{-3}. Beryllium-7 (half-life of $T_{1/2}=53.2$ days) is a cosmogenic radionuclide, which is continuously produced in the atmosphere. About 67% of Be-7 is produced in the stratosphere and 33% in the troposphere [4]. That is why its presence in the near-surface atmospheric layer should have some affinity with the stratospheric radiocaesium component. On the other hand, caesium present in the above layer comes from the biological material and soil having been lifted from the soil surface.

Table 1. Mean annual Cs-137 activity concentrations for the region of Białystok in 1992–1999.

Year	Mean concentration Cs-137 (μBq m^{-3})	Effective dose equivalent (pSv)
1992	5.3±4.3	1509±1224
1993	4.7±6.5	1338±1851
1994	3.2±2.2	911±626
1995	3.4±2.2	968±626
1996	3.2±2.2	911±626
1997	2.2±1.5	626±427
1998	2.1±1.7	598±484
1999	2.2±1.5	626±427

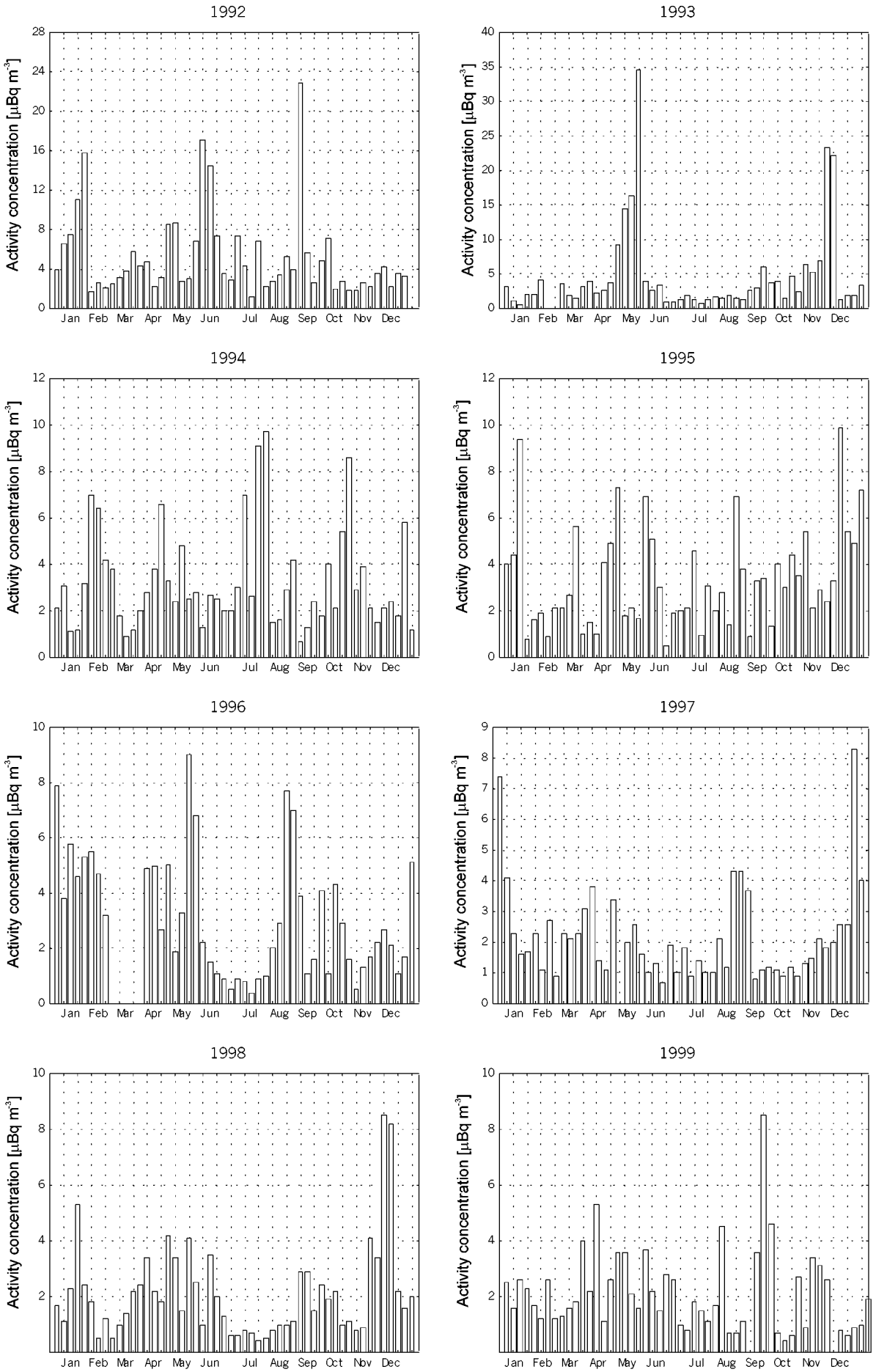

Fig. 1. Weekly changes in Cs-137 activity concentrations in annual cycles in 1992–1999.

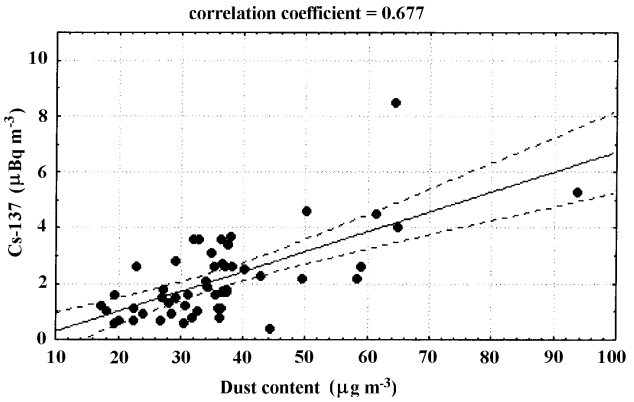

Fig. 2. Correlation between Cs-137 and dust content in 1999.

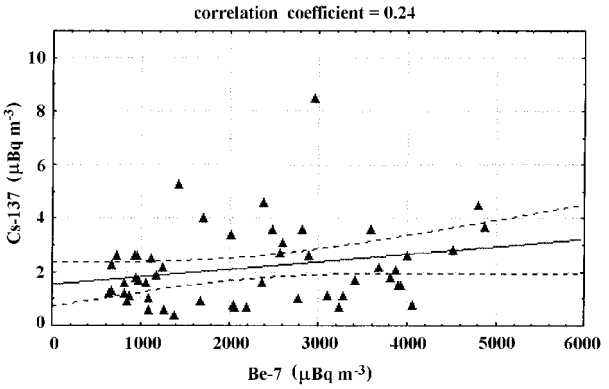

Fig. 3. Dependence between Cs-137 and Be-7 in 1999.

The strong correlation between Cs-137 and the dust content in air (Pearson's correlation coefficient r=0.677) and the weak correlation between Cs-137 and Be-7 (Pearson's correlation coefficient r=0.24) determined in 1999, indicate that at present, the prevailing pathway leading to the radiocaesium presence in air is its re-suspension from the contaminated soil. This fact explains the great variation in airborne radiocaesium levels in the annual cycle. The local resuspension of Cs-137 is affected by the following factors: wind velocity, intensity and frequency of rainfall, kind and structure of soil, as well as the humidity of the soil [10].

The analysis of Cs-137 concentrations in the annual cycle in 1992–1999 reveals that the maximum values exceed the annual averages by a factor of 2.8 to 7.3. Increased spring values may be due to the fact that soil dries up after winter, which facilitates its transport by winds. It can be also connected with the fact that fine fragments of dry plants are transported by the wind in the vicinity of the station after the haymaking. This conclusion has been drawn from many years of observations of agricultural procedures taking place around the station. The first hay harvest in May and the second one in August may be responsible for two Cs-137 peaks in summer, which have been found in Bavaria near Munich [14], although no explanation was provided for this fact. Higher Cs-137 levels are also observed in winter months, which cannot be accounted for by local re-suspension since this effect may be limited by snow covering the frozen ground. However, during winters without frost or snow, soil particles may also be lifted up in air [5], which may be due to increased dust content in air caused by dust emission from thermal-electric power stations during the heating season.

What should specially be noted here are higher airborne radiocaesium levels detected in May of 1993 (maximum value of 35.5 μBq m^{-3}), which may be accounted for by very strong spruce pollination at that time. A similar effect has already been observed in the studies carried out in Bavaria [1].

In addition, over the period of 8 years of measurements two peaks exceeding the concentration of 20 μBq m^{-3} were detected in 1992 and 1993. This may be due to the big forest fires spreading in Belarus in August 1993 and the long period of eastern air circulation by the end of 1993, as a result of which large amounts of air were transported to Poland from the heavily contaminated regions of Belarus and Ukraine [6].

The effect of radiation hazard from inhalation is quite negligible (626 pSv in 1999), as it represents only a thousandth of the percentage of the total effective dose equivalent from the environmental caesium contamination (value 2.9 mSv [7]). However, the analysis of the above problem is of practical importance since it involves radiological monitoring of the environment. The results of the analysis help to better understand and interpret a possible emergency situation of increased caesium concentrations since they point to the natural factors that cause variation in air contamination.

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