TEMPORAL VARIATIONS OF ^{137}Cs IN SURFACE AIR IN BRATISLAVA, SLOVAKIA, OVER 33 YEARS

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Abstract. The objective of this study is to evaluate the changes in surface atmospheric distribution of ^{137}Cs before and after the Chernobyl accident. Measurements of ^{137}Cs in the air carried out in Bratislava, Slovak Republic, from 1977 until 2010 years show that the main source of ^{137}Cs surface air activity is the residue of the global fallout from the atmospheric nuclear weapon tests. From these measurements it follows that the decrease of ^{137}Cs concentration in air has an exponential dependence with the ecological half-life τ = 59.1 months, except for a significant increase in activity level during the years 1986–1988 due to the Chernobyl accident. At present the level of airborne ^{137}Cs activity is about 0.3 μBq/m^3. It appears that the decreasing trend (with ecological half-life) has stopped after the year 2008, and a shift of the ^{137}Cs concentration peak from summer to winter season is observed. This effect may be associated with transfer of radiocaesium from soil to vegetation and subsequent burning of biomass, or with soil resuspension.

Keywords: ^{137}Cs in surface air, Long-term variation, seasonal variation, Chernobyl fallout; nuclear weapons fallout

Introduction

The radionuclide ^{137}Cs is released to the environment by several types of nuclear activities including testing of nuclear weapons and accidents in nuclear facilities, operation of nuclear power reactors, and reprocessing of spent nuclear fuel. The ^{137}Cs release into atmosphere is a result of nuclear fission, and it can be considered as one of the most hazardous radionuclides in the environment. It has a high fission yield, long physical half-life (30.07 years), high solubility and physico-chemical properties similar to potassium.

The testing of nuclear weapons in the atmosphere involved unrestrained releases of radioactive materials directly to the environment. During atmospheric nuclear bomb testing ^{137}Cs was released in the stratosphere. Approximately 10·PBq **(**1 PBq=10^{15} Bq) of ^{137}Cs, was injected in the stratosphere (UNSCEAR, 1982) from past tests. A major part of the emissions

(~60%) took place during the period 1961–1962. One may expect that the peak concentration of ^{137}Cs in surface air from nuclear bomb tests was observed in 1963 when the radioactive fission products from the explosions began to fall down from the stratospheric reservoir into ground surface atmosphere.

By exchange processes ^{137}Cs was transported to the troposphere and further as fallout to the Earth surface. After the end of the atmospheric nuclear weapon tests in 1963 (thermonuclear explosions in the atmosphere were completely terminated in 1980), the content of this radionuclide in the environmental reservoirs gradually decreased**.** Until the Chernobyl event on 26 April 1986, surface air activity of ^{137}Cs occurred mostly as a result of global fallout from atmospheric nuclear weapons tests. The Chernobyl event was restricted in space and time. The radionuclide releases from the damaged reactor occurred mainly over a 10-day period *(United Nations, 1994).* Totally 77 PBq of ^{137}Cs entered the atmosphere within a few days following the accident. The localized fallout from the event covers large areas of Europe, including Slovakia. According to the assessment of the Russian National Report prepared by the Institute of Nuclear Energy Safety, Moscow, 0.18 PBq of ^{137}Cs was deposited to the territory of Slovakia, which is 0.28% of the total deposition in Europe (Shoigu et al., 2008). The distribution of deposition was non-even. Based on measuremenst of ^{137}Cs activity over the whole territory of Slovakia from 1990 till 2003 by means of ground gamma-spectrometers and atmospheric aerosols sampling equipment it was established (Gluch, 2006) that the generally highest values (> 3000 Bq/m^2) were observed in the areas with altitude exceeding 800 m (The HighTatras, The Low Tatras, Štavnica vrchy). The maximum measured values occurred in the vicinity of the towns Banska Štiavnica (18000 Bq.m^{-2}) and Novy Tekov (28700 Bq.m^{-2}). In the vicinity of Bratislava the activity was in the range of 1500–1800 Bq/m^2 (with reference to 1.1.2005).

Systematic monitoring of airborne ^{137}Cs activity has been done by the Department of Nuclear Physics and Biophysics of the Comenius University in Slovakia over the periods 1977–1987 and 2002–2010. In the atmosphere the concentrations of ^{137}Cs (together with ^{7}Be, ^{40}K and ^{210}Pb, were measured in industrial city of Bratislava. Bratislava (with 0.5 million inhabitants) is situated near borders with Hungary, Austria and Czech Republic and at a distance of 1000 km from the Chernobyl Power Plant, where the 1986 accident occurred.

Material and Methods

Aerosol particles in the atmosphere were collected using aerosol filters SYMPOR 3 (1977-1988) and nitro-cellulose filters (2002-2010), both with a collection efficiency of approximately 100 %. The sampling location was at the Meteorological Station near the Faculty of Mathematics, Physics and Informatics (FMPI), Comenius University, Bratislava (48° 9′ N, 17 ° 7′ E, 164 m a.s.l.). Using a sampler device with an air-flow rate of 30 m^3.h^{-1} aerosol particles were collected on the nitro-cellulose membrane filters (PRAGOPOR, pore size 0.85 μm, the collection efficiency approximately 100 %). The sampler device is situated at height of 2.85 m above ground at the Meteorological Station near the FMPI. The filters are

changed every week and about 3000 m^3 of air is pumped through each sample. In September 2004 the new sampling device was launched and the volume of pumped air increased twofold.

Gamma spectrometry on the air-filter samples was performed in the low-level background counting laboratory of the Department of Nuclear Physics and Biophysics of the Comenius University in Bratislava, Slovakia, using a ORTEC HPGe detector with Be window and Canberra HPGe detector (177 cm^3) with a carbon window, placed in a low-level background shield. The measuring time was 24 hours or more. The peaks corresponding to the 46.5 keV, 477 keV, 662 keV and 1461 keV gamma rays of ^{210}Pb, ^{7}Be, ^{137}Cs and ^{40}K, respectively, were recorded for activity determination. The uncertainty in the results was mainly due to counting statistics, which was normally 3% or better. The count-rates in the full-energy peaks were corrected for the background of the measurement system and for self-absorption. The detection efficiency for the sample geometry was evaluated from a Monte Carlo model using the GEANT 3 code.

In our department concentration of ^{137}Cs was measured from July 01, 1977 (*Povinec et al., 1988)* up to present, regrettably with an interval from August 1, 1988 until January 1, 2002. During the years 1993 - 2000 regular collection of aerosol samples for routine environmental air monitoring was carried out in six measuring points within the Radiation Monitoring Network of Slovak Republik (*Cabanekova, 1998; Cabanekova and Gomola, 2001).*

Result and discussion

a. Long-term variations

The overall results of the air-particulate monitoring for ^{137}Cs is shown in Figure 1 and covers a period from July 01, 1977 (*Povinec et al., 1988)* up to present. In addition to our date we included averaged values from 1993-2000 in the six measuring points used by The Radiation Monitoring Network of Slovak Republic (*Cabanekova, Vladar, 1998, Cabanekova, Gomola, 2001)***.**

In Bratislava the annual average activity of ^{137}Cs in surface air decreased regularly from 130 μBq/m^3 in 1977 down to 0.3 μBq/m^3 in 2009, except for a significant increase during the years 1986–1987 due to the Chernobyl accident.

From these measurements it appears that the decrease in concentration of ^{137}Cs in air follows an exponential trend according to the following equation $A = 130 \exp(-0.0169\ \mathbf{t})$, if **t** is expressed in months after 1.01.1977. The apparent ecological mean life-time is 59.1 months (the corresponding half-life time is around 41 months).

Before the Chernobyl accident, the concentration of ^{137}Cs decreased with approximately the same effective life-time as after 1993 (Fig. 1). In 1983 it varied within the range 20–30 μBq/m^3. Measurements performed in Sweden (*Kulan, 2006*) gave a value of 28.8 μBq/m^3 during the period August 1972 - December 1985, close to the present data.

In Slovakia the maximum air concentration of ^{137}Cs from the Chernobyl accident at the ground level was registered on May 1, 1986. Monthly averaged concentration for May 1986 was 14500 μBq/m^3 (*Povinec et al., 1988)*. Then it decreased with an ecological life-time of 2.8 months (half-time 1.93 month). At the end of 1987 it approached again the level determined by the previous recorded decline in global fallout (see inserted picture in Fig. 1).

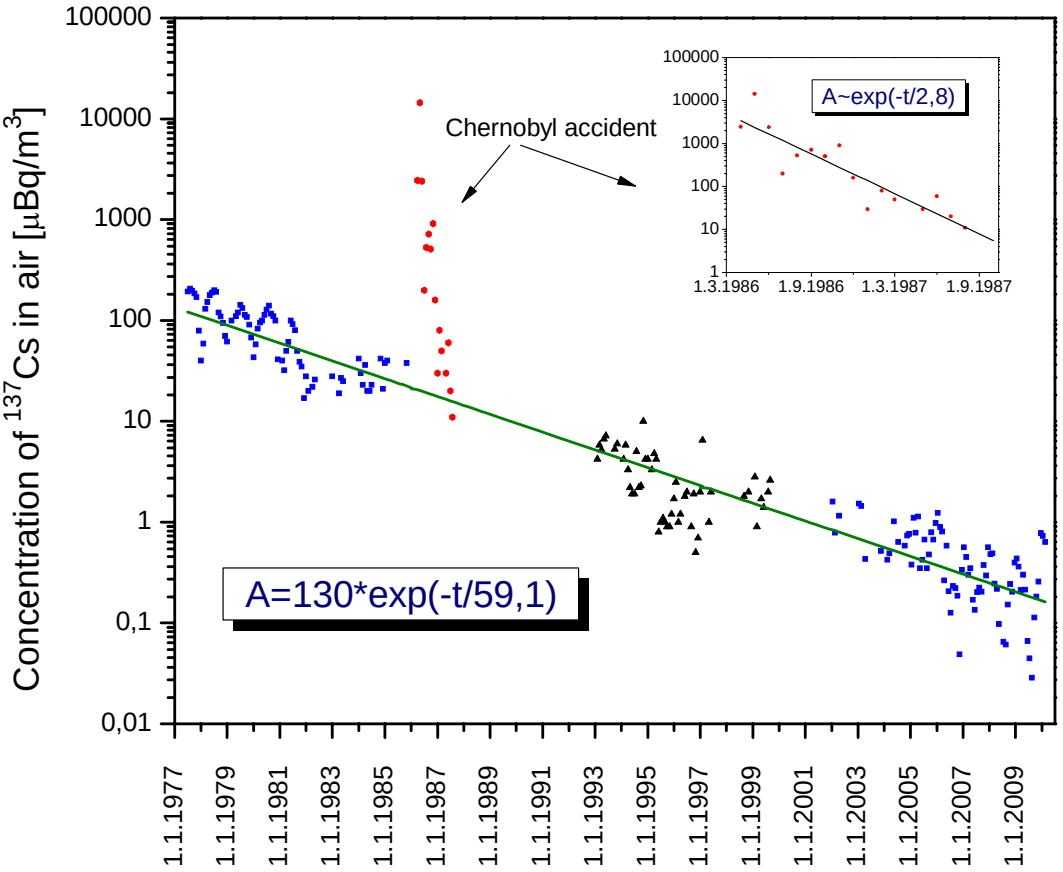

Fig.1. Long-term variation of ^{137}Cs in surface air in Bratislava, based on monthly mean values. Inserted picture is monthly average values during 1986–1987. The time **t** in the formula is in months.

The most active years of testing from the standpoint of the total explosive yields were in 1961–1962. Subsequently, the maximum concentration of ^{137}Cs in surface air was observed in 1963 *(Makhonko, Kim, 2002)*. They obtained a value of 2450 μBq/m^3 as a weighted mean over 89 sites used to monitor radioactive aerosols localized in the territory of the former USSR. If to extrapolate our data back to 1963**,** the concentration of ^{137}Cs in the vicinity of Bratislava was 2220 μBq/m^3 at that time.

The trend of decreasing volume activity of ^{137}Cs in air appears to cease after the year of 2007 (Fig. 2). This indicates that the current main source of atmospheric ^{137}Cs in Slovakia is different from the previous period. As demonstrated by analysis of moss samples

(Aleksiayenak et al., 2011) there is still a considerable residual ^{137}Cs activity level in terrestrial surfaces affected by fallout from the Chernobyl accident, and it is conceivable that release of particulate material from these surfaces could be a main source of the current level of ^{137}Cs in air in Slovakia.

b. Seasonal variations

In recent years, in contrast to the period 1977–1980, we observe a shift of maximum concentration of ^{137}Cs from summer season to winter season (Fig. 2). This is another indication of a change of source distribution of ^{137}Cs in air.

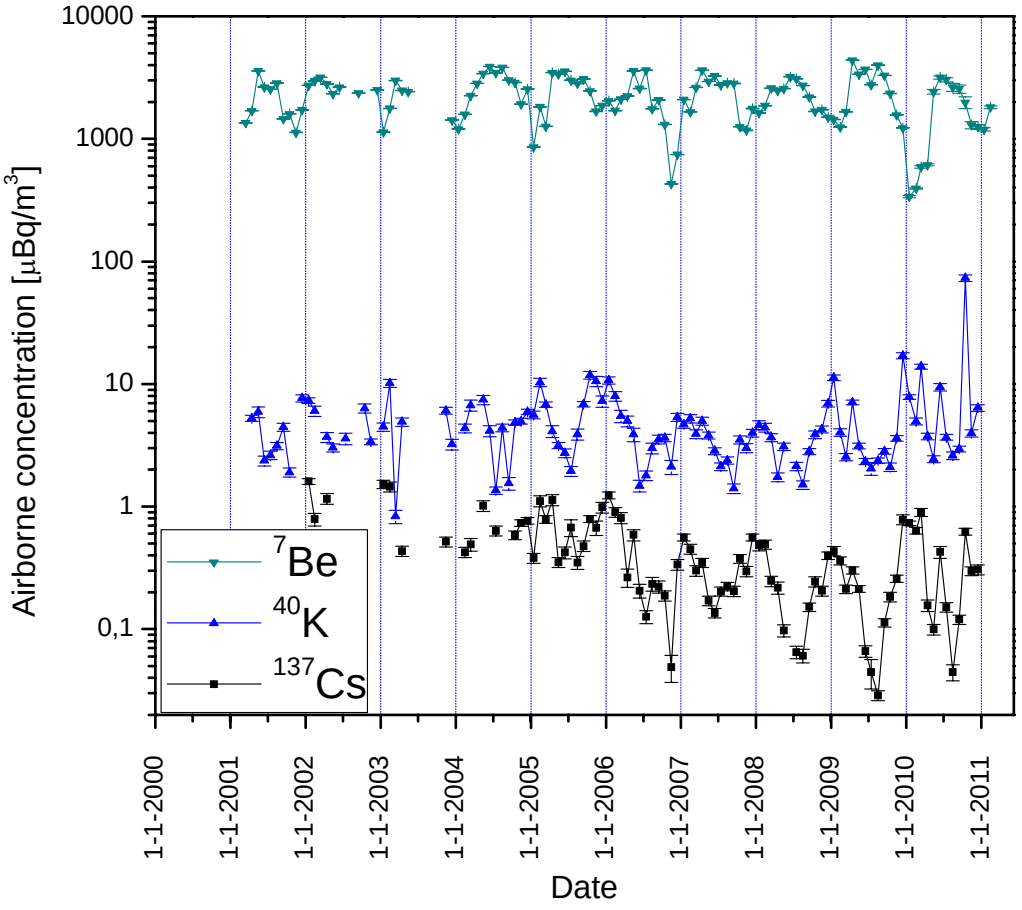

Fig.2. Seasonal variation airborne concentration of ^{137}Cs and ^{40}K (date with standard deviation) in air in Bratislava over the last years. For comparison the seasonal variations of ^{7}Be concentrations are shown.

The seasonal variation of ^{137}Cs activity concentration in surface air correlates well (R = 0.78) with that of ^{40}K (Fig. 3). This has been expected as both the source-term (a resuspension from soil) and behavior are similar. We included in this evaluation only the data for the 2007-2010 period, as we assumed that the change in ^{137}Cs activity concentration in the atmosphere ceased in 2007. The average ^{40}K and ^{137}Cs activity concentrations for this period are 4.4 µBq m^{-3} and 0.3 µBq m^{-3}, respectively (the ^{137}Cs/^{40}K activity ratio is 0.069).

^{137}Cs and ^{40}K levels measured in oak and beech leaves collected from different regions of Slovakia in 2001 and 2002 varied within the range of 0.4–3.5 Bq kg^{-1} (mean value 1.73 Bq kg^{-1}) and 173–365 Bq kg^{-1} (mean value 230 Bq/kg), respectively, Table 1. However, ^{137}Cs activities in conifer needles were below a detection limit. This would indicate that a root uptake of ^{137}Cs by plants could be a dominant process. The concentration of stable K and Cs were determined in Frank Laboratory of Neutron Physics, JINR, at reactor IBR-2 using INAA method.

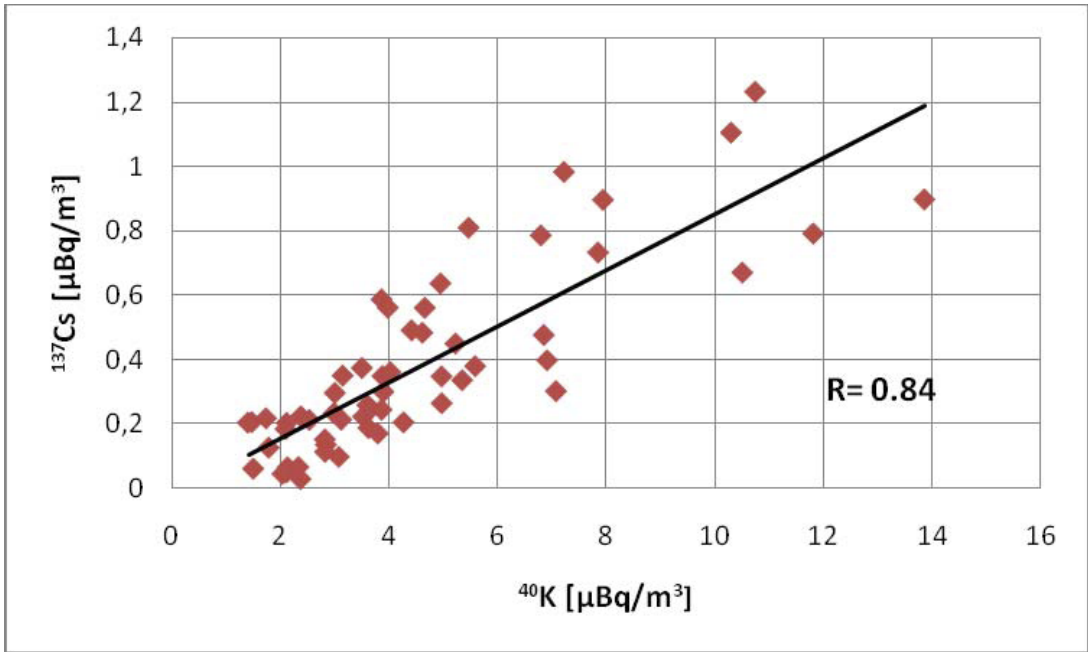

Fig.3. A plot of correlation between activity of ^{137}Cs and ^{40}K in surface air (Bratislava). Only data from the period 2007-2010 are included.

In the foliage samples the ^{137}Cs/^{40}K activity ratio is 0.01. Higher ratios (0.064) were registered by Ko et al. (2002) in broadleaf tree litter. Even higher levels of ^{137}Cs were registered in piedmont broadleaved forests Malyi Utrish (44^{0}44´N, 37^{0}26´E) in Russia (approximately the same latitude as Bratislava). Pokarzhevskii et al. (2003) reported ^{137}Cs levels between 76 and 140 Bq kg^{-1} in litter found below oak, hornbeam and beech trees.

Table 1. Concentration of radionuclides in Slovakian samples of foliages, needles and moss.

Samples	Number of samples	Stable K, mg/kg	^{40}K, Bq/kg	Stable Cs, mg/kg	^{137}Cs, Bq/kg
Foliage (oak, beech)	25	9840	230	0.044	1,77
Spruce needles	2	6360	120	0.22	<0,2 (DL)
Moss	11	7080	138	0.41	30

Possible reasons of increased atmospheric ^{137}Cs levels during the autumn–winter season may be due to:

(i) surface soil resuspension and transport of released ^{137}Cs by winds, particularly from open agricultural areas;

(ii) releases of decomposed plant materials by wind erosion;

(iii) burning of biomass (Bourcier et al., 2010);

(iv) specific meteorological conditions during winter with temperature gradient inversions (Povinec et al., 2011).

All these mechanisms could contribute significantly to the observed recent changes in the seasonality of ^{137}Cs in Slovakia, where the alternation of annual seasons is regular, and the period of snow cover is insignificant. Bratislava with 0.5 million of inhabitants is also a large industrial zone where specific meteorological conditions during winter prevent dispersion of pollutants from the town, as documented by $^{14}CO_2$ observations in the ground-level air (Povinec et al., 2011). The fact that the $^{137}Cs/^{40}K$ ratio observed in aerosols (cf. Fig. 3) exceeds recent values observed in vegetation may indicate that soil resuspension is responsible for at least part of the increased ^{137}Cs activity observed during the winter season. The mean ^{137}Cs (15 Bq kg^{-1}) and ^{40}K (300) levels observed in soil around Bratislava gave for the $^{137}Cs/^{40}K$ activity ratio a mean value of 0.05, which is closer to the value for the surface air (0.07) than for the tree leaves (0.01). A few single ^{137}Cs peaks observed in one-two months (Fig. 2) may indicate contributions either from biomass burning of highly contaminated areas in the Eastern Europe (Povinec et al., 2011), or from Saharan dust events (Pham et al., 2005).

Conclusions

Sources and variations of ^{137}Cs and ^{40}K in the ground-level air of Bratislava have been compared and discussed. The main findings of the paper may be summarised as follows:

(i) The ^{137}Cs activity concentration in the surface air between 1977 and 2010 has been decreasing with an ecological half-life of 3.4 years (high values observed during 1986 and 1987 due to the Chernobyl accident were excluded from the evaluation). However, during 2007-2010 the yearly averaged ^{137}Cs activity concentrations were almost the same.

(ii) The increased atmospheric ^{137}Cs and ^{40}K levels observed during the autumn-winter season may be due to surface soil resuspension and radionuclide transport by winds, particularly from open agricultural areas (also confirmed by high correlation coefficient, R = 0.84, between the ^{137}Cs and ^{40}K atmospheric levels). Decomposed plant materials, biomass burning and specific meteorological conditions during winter with temperature gradient inversions could also contribute to higher ^{137}Cs and ^{40}K levels observed during the autumn and winter months.

(iii) The $^{137}Cs/^{40}K$ activity ratio for the surface air (0.07) is closer to the mean value observed in soil (0.05) than to the mean value for the tree leaves (0.01), what would also indicate a predominance of soil resuspension processes on the atmospheric concentrations of these radionuclides during the winter season.

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