E14-2011-85

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VARIATIONS OF ^{137}Cs AND ^{40}K IN THE SURFACE AIR OF BRATISLAVA (SLOVAKIA) — INDICATIONS OF SOIL RESUSPENSION PROCESSES

Submitted to «Atmospheric Environment»

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Сикора И. и др. E4-2011-85
Вариации ^{137}Cs и ^{40}K в приземном слое воздуха в Братиславе (Словакия) — указание на процессы ресуспензии почвы

В работе приводятся и обсуждаются источники ^{137}Cs и ^{40}K и их вариации в приземном воздухе Братиславы. Концентрация активности ^{137}Cs в приземном воздухе Братиславы в период между 1977 и 2007 гг. снижалась с экологическим периодом полураспада 3,4 года (высокие значения, наблюдавшиеся в 1986 и 1987 гг. из-за аварии на Чернобыльской АЭС, в оценку не входили). Однако в 2007–2010 гг. ежегодные усредненные концентрации ^{137}Cs были почти постоянными. Повышенный уровень^{137}Cs и ^{40}K в атмосфере, наблюдаемый в течение зимы, может быть связан с ресуспензией поверхностнго слоя почвы и переносом радионуклидов ветрами, особенно с открытых сельскохозяйственных территорий (что также подтверждается высоким коэффициентом корреляции атмосферных концентраций ^{137}Cs и ^{40}K: $R = 0{,}84$). Отношение активности^{137}Cs/^{40}K в приземном слое воздуха (0,07) ближе к среднему значению в почве (0,05), чем к среднему значению в листьях деревьев (0,01), что также указывает на преобладающее влияние почвенных процессов ресуспензии на атмосферные концентрации этих радионуклидов в зимний период.

Работа выполнена на кафедре ядерной физики и биофизики факультета математики, физики и информатики Университета им. Я. А. Коменского, Братислава, Словакия, и в Лаборатории нейтронной физики им. И. М. Франка ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2011

Sýkora I. et al. E4-2011-85
Variations of ^{137}Cs and ^{40}K in the Surface Air of Bratislava (Slovakia) — Indications of Soil Resuspension Processes

Sources and variations of ^{137}Cs and ^{40}K in the ground-level air of Bratislava are presented and discussed. The ^{137}Cs activity concentration in the surface air between 1977 and 2007 was 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 constant. The increased atmospheric ^{137}Cs and ^{40}K levels observed during winter may be due to surface soil resuspension and radionuclide transport by winds, particularly from open agricultural areas (also confirmed by high correlation coefficient between the ^{137}Cs and ^{40}K atmospheric concentrations, $R = 0.84$). 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 observed in tree leaves (0.01), which would also indicate a predominant influence of soil resuspension processes on the atmospheric concentrations of these radionuclides during winter.

The investigation was performed at the Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University, Bratislava, Slovakia, and at the Frank Laboratory of Neutron Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2011

INTRODUCTION

The anthropogenic ^{137}Cs has been released to the environment by various nuclear activities including atmospheric testing of nuclear weapons, accidents in nuclear facilities, reprocessing of spent nuclear fuel, and operation of nuclear power reactors (Livingston and Povinec, 2000). ^{137}Cs originates in nuclear fission, and it has been considered as one of the most hazardous radionuclide found in the environment. It has a high fission yield, long physical half-life (30.07 years), high solubility and physico-chemical properties similar to those of potassium.

The testing of nuclear weapons in the atmosphere involved unrestrained releases of radioactive materials directly to the atmosphere. During atmospheric nuclear bomb testing ^{137}Cs was transported to the upper atmosphere, including the stratosphere. Approximately 10 PBq of ^{137}Cs was injected into the stratosphere from nuclear weapons tests (UNSCEAR, 1982, 1994). A major part of the emissions ($\sim 60\%$) took place during the period 1961–1962 when nuclear weapons tests were carried out mostly at Novaya Zemlya Island in the Kara Sea.

After the moratorium on atmospheric nuclear weapons tests signed in 1963, the new supply of bomb-produced radionuclides to the stratosphere was limited, as contributions from nuclear tests carried out by other countries was almost negligible, although nuclear explosions in the atmosphere were terminated only in 1980. Therefore, the peak concentration of ^{137}Cs in the surface air was observed in 1963 when the radioactive fission products were transported from the stratosphere to the troposphere, and further as global fallout to the earth surface with gradually decreasing levels (Livingston and Povinec, 2002). Makhonko and Kim (2002) reported for the maximum ^{137}Cs surface air activity concentration measured in 1963 at 89 radioactive aerosols monitoring stations localized over the territory of the former USSR a mean value of 2450 $\mu\beta\varsigma$ m^{-3}.

Therefore, until the Chernobyl event, which occurred on April 26, 1986, the ^{137}Cs activity concentration in surface air was mostly due to global fallout from atmospheric nuclear weapons tests. The Chernobyl event was, however, restricted in space and time (IAEA, 2006). The radionuclide releases from the damaged reactor occurred mainly over a 10-day period when about 85 PBq of ^{137}Cs entered the troposphere (UNSCEAR, 2008). The radionuclide fallout from the event has

covered large areas of Europe, including Slovakia (Povinec et al., 1988). Shoigu and Bolshov (2008) estimated that about 0.18 PBq of ^{137}Cs was deposited over the territory of Slovakia, representing 0.28% of the total deposition in Europe. The distribution of deposition was non-even. It was established (Gluch et al., 2005) that the highest ^{137}Cs deposition (> 3000 Bq $\cdot$ m^{-2}) was observed in areas with altitudes exceeding 800 m (The High Tatras, The Low Tatras, Štavnické vrchy). The maximum values were measured in the vicinity of the towns of Novy Tekov (28700 Bq $\cdot$ m^{-2}) and Banska Štiavnica (18000 Bq $\cdot$ m^{-2}). In the vicinity of Bratislava the surface contamination was in the range of 1500–1800 Bq $\cdot$ m^{-2} (with reference to 1.1.2005).

Monitoring of airborne ^{137}Cs activity (together with ^{7}Be, ^{40}K and ^{210}Pb) has been carried out by the Department of Nuclear Physics and Biophysics of the Comenius University in Slovakia over the periods 1976–1993 (Povinec et al., 1988), and after 2000, when large volume air sampling became available. During the years 1993–2000 a regular collection of aerosol samples for routine environmental air monitoring was carried out within the Radiation Monitoring Network of the Slovak Republic at six monitoring stations (Cabáneková, 1998; Cabáneková and Gomola, 2001).

MATERIALS AND METHODS

Aerosol particles in the atmosphere were collected using aerosol filters SYMPOR 3 (1976–1993) and nitro-cellulose membrane filters PRAGOPOR, pore size 0.85 μm (2000–2010). Both types of filters had a collection efficiency of approximately 100%. The sampling location was located at the Slovak Meteorological Institute at Koliba, Bratislava (1977–1993), and from 2000 at the Meteorological Station of the Faculty of Mathematics, Physics and Informatics of the Comenius University at Mlynská dolina ($48^\circ\,9'$ N, $17^\circ\,7'$ E, 164 m a.s.l.). The sampling device has been situated at a height of 2.8 m above the ground. The air-flow rate has been 30 m$^3 \cdot$ h^{-1}. The filters were changed every week, so about 3000 m of air was pumped through each sample. In September 2004 a new sampling device was launched, and the volume of pumped air increased twofold.

Gamma spectrometry on the air-filter samples was carried out in the low-level gamma-spectrometry laboratory of the Department of Nuclear Physics and Biophysics using an ORTEC planar HPGe detector with Be window, and a Canberra coaxial HPGe detector (177 cm^3) with a carbon window, placed in low-level background shields. The measuring time was 24 hours or more. The peaks corresponding to the 46.5, 477, 662 and 1461 keV gamma rays of ^{210}Pb, ^{7}Be, ^{137}Cs and ^{40}K, respectively, have been used for activity determination (data on ^{210}Pb and ^{7}Be will be reported in a separate paper). The results uncertainties were mainly due to counting statistics, which was around 3% or better. The

count-rates in the full-energy peaks were corrected for the background of the counting system and for self-absorption (Sýkora et al., 2008). The detection efficiency for the sample geometry was evaluated from a Monte Carlo model using a GEANT 3 code.

RESULT AND DISCUSSION

Long-Term Cs Variations. The results of ^{137}Cs monitoring of aerosols in the Bratislava air between 1977 and 2010 are shown in Fig. 1. In addition to the data obtained in the Department of Nuclear Physics and Biophysics, we also included averaged ^{137}Cs values for the period 1993–2000 reported by the Radiation Monitoring Network of the Slovak Republic (Cabáneková, 1998; Cabáneková and Gomola, 2001). The annual average ^{137}Cs activity concentration in the Bratislava surface air has been decreasing regularly from 130 μBq $\cdot$ m^{-3} in 1977 down

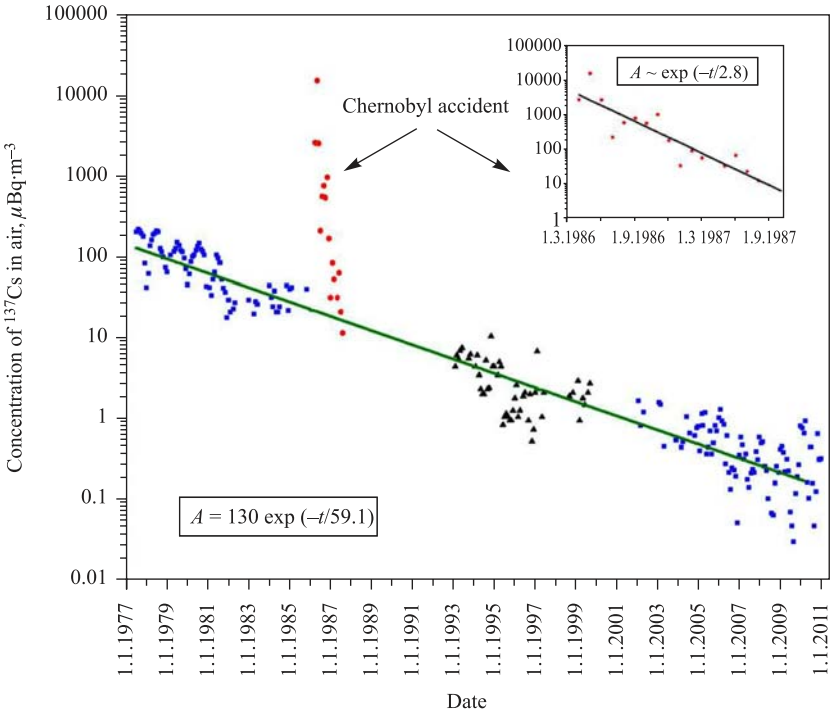

Fig. 1. Long-term variations of ^{137}Cs activity concentrations in the Bratislava surface air, based on monthly mean values (the inserted picture represents monthly average values during 1986–1987)

to 0.3 $\mu\mathrm{Bq}\cdot\mathrm{m}^{-3}$ in 2009, except for a significant increase observed during the years 1986–1987 due to the Chernobyl accident. The observed decrease in the ^{137}Cs activity concentration in the air follows an exponential trend $A = 130\ \exp(-0.0169t)$, (where t is expressed in months) with an apparent ecological mean lifetime of 59 months, and the corresponding half-life of 41 months (3.4 years). If we extrapolate our data back to 1963, the annual average ^{137}Cs activity concentration in the Bratislava air would be 2220 $\mu\mathrm{Bq}\cdot\mathrm{m}^{-3}$. During the eighties, just before the Chernobyl accident, the ^{137}Cs activity concentration in the Bratislava air varied within the range of 20–30 $\mu\mathrm{Bq}\cdot\mathrm{m}^{-3}$, similarly as in Sweden, where Kulan (2006) reported for the period August 1972–December 1985 an average value of 28.8 $\mu\mathrm{Bq}\cdot\mathrm{m}^{-3}$.

The ^{137}Cs activity concentration before the Chernobyl accident was decreasing with approximately the same effective half-life as after 1993 (Fig. 1). The maximum ^{137}Cs activity concentration in the Bratislava surface air due to the Chernobyl accident was registered on April 30, 1986 (Bratislava is around 1000 km far from Chernobyl). The monthly averaged concentration for May

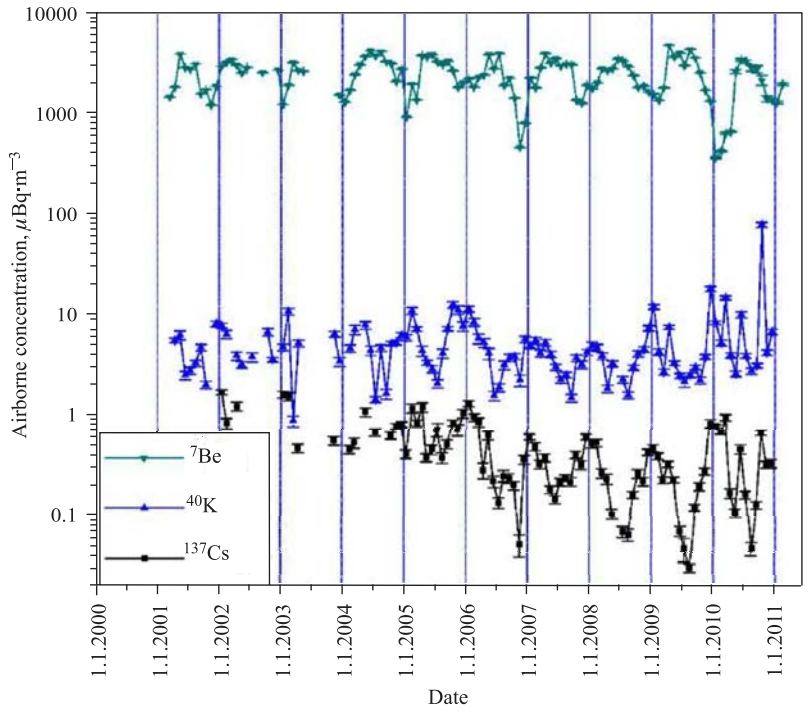

Fig. 2. Seasonal variations of airborne activity concentrations of Be, Cs, and K (with standard deviations) in the Bratislava air during the last decade

1986 was 14500 μBq $\cdot$ m^{-3} (Povinec et al., 1988). Then it decreased with an ecological lifetime of 2.8 months, and at the end of 1987 it approached again the level determined by the previous recorded decline in global fallout (see the inserted figure in Fig. 1).

The trend of decreasing ^{137}Cs activity concentration in surface air appears to cease after 2007 (Fig. 2). This would indicate that the current main source of atmospheric ^{137}Cs in Slovakia may be different from the previous period. The explanation may be in the fact that the higher soil erosion during winter and corresponding release of small particles carrying ^{137}Cs are due to the fact that agricultural fields are not any longer covered by vegetation after the harvest of crops, and will stay like that until the next spring, when the new crop starts growing. In addition, stronger winds during the winter seasons may play a role.

Seasonal ^{137}Cs and ^{40}K Variations. Seasonal variations of airborne concentrations of ^{137}Cs, ^{40}K and ^{7}Be in the Bratislava air during the last decade are compared in Fig. 2. In contrast to the pre-Chernobyl period we have observed a shift of maximum ^{137}Cs activity concentration from the summer season to the winter season. This is due to changes in sources of anthropogenic radionuclides in the surface air.

During the pre-Chernobyl period the main source of global fallout radionuclides in the surface air was stratospheric radioactive air. Due to specific exchange processes between the stratosphere and the troposphere, especially during the late spring and early summer when rising hot air provokes the descent of cold air masses from the lower stratosphere, enhanced concentrations of radionuclides were observed in the lower troposphere, causing spring and early summer ^{137}Cs maxima in the surface air.

During the post-Chernobyl period the stratospheric reservoir of ^{137}Cs has not been anymore its dominant source, but a resuspension of global and Chernobyl fallout ^{137}Cs from soil has become important source of ^{137}Cs in the surface air. The typical spring–summer ^{137}Cs maxima in the surface air were not anymore observed during the early eighties, just before the Chernobyl accident in 1986 (Fig. 1), but they were replaced by winter maxima as seen in Fig. 2.

Much of the ^{137}Cs activity previously deposited over Slovakia still resides in the surface horizon of the soil (Daniel et al., 1996). As demonstrated by analysis of moss samples (Aleksiayenak et al., 2011), there is still a residual ^{137}Cs on the Earth surface affected by the Chernobyl accident. Ko et al. (2002) found that the ^{137}Cs levels in the soil have been decreasing with depth, while stable Cs and ^{40}K were almost constant. A larger fraction of ^{137}Cs is still in the upper layer of soil (up to 10 cm). Part of the ^{137}Cs may occur in forms available for root uptake by plants, which is largely recycled annually through leaching and litterfall.

The ^{40}K record presented in Fig. 2 for the last decade is similar to the one for ^{137}Cs, showing concentration maxima during the winter, documenting similar origin and behavior of these radionuclides in the surface air. The main source of

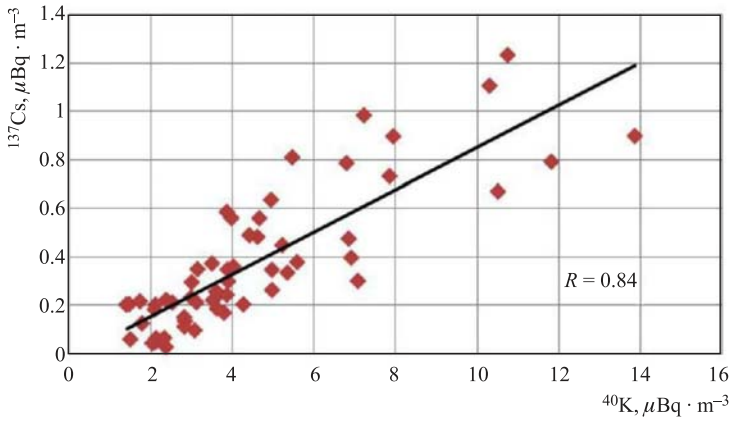

Fig. 3. Correlation between the ^{137}Cs and ^{40}K activity concentrations in the Bratislava surface air (only data from the period 2007–2010 were included in the evaluation)

^{40}K in the air is its transport by wind from the soil, as documented by similar yearly averaged ^{40}K levels in the air. The ^{7}Be record, also presented in Fig. 2 for comparison, shows, however, a different structure, confirming its cosmogenic origin with maxima observed in summer due to transport of ^{7}Be from the lower stratosphere and the upper troposphere to the ground-level air.

The seasonal variation of ^{137}Cs activity concentration in surface air correlates well with that of ^{40}K, the correlation coefficient $R = 0.84$ (Fig. 3). This has been expected as for both radionuclides the source-term (a resuspension from soil) and their behavior in the atmosphere are similar. We included in this evaluation only the data for 2007–2010, 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 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 1.77 Bq · kg^{-1}) and 173–365 Bq · kg^{-1} (mean value 230 Bq · kg^{-1}), respectively (see table). The ^{40}K levels in conifer needles were $\sim$ 120 Bq · kg^{-1}, however, the ^{137}Cs levels were below the detection limit (0.2 Bq · kg^{-1}). This would indicate that a root uptake of ^{137}Cs by plants could be a dominant process.

The ^{137}Cs/^{40}K activity ratio measured in foliage samples is 0.01. Higher ^{137}Cs/^{40}K ratios (0.064) were measured by Ko et al. (2002) in broadleaf tree litter. Higher ^{137}Cs levels (between 76 and 140 Bq · kg^{-1}) were also measured by Pokarzhevskii et al. (2003) in litter found below oaks, hornbeam and beech

Mean ^{40}K and ^{137}Cs levels in foliages, needles and moss samples collected in Slovakia (2001–2002)

Samples	Number of samples	K*, $mg \cdot kg^{-1}$	^{40}K, $Bq \cdot kg^{-1}$	Cs*, $mg \cdot kg^{-1}$	^{137}Cs, $Bq \cdot kg^{-1}$
Foliage (oak, beech)	25	9840	230	0.044	1.77
Spruce needles	2	6360	120	0.22	< 0.2
Moss	11	7080	138	0.41	30

*Concentrations of stable K and Cs were determined in the FLNP, JINR, using the INAA.

trees in piedmont broadleaved forests of Malyi Utrish ($44^\circ\,44'$ N, $37^\circ\,26'$ E) in Russia (approximately of the same latitude as Bratislava).

Possible reasons of observed 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);

(v) Saharan dust events (Pham et al., 2005).

All these mechanisms could contribute 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 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 exceeds recent values observed in vegetation may indicate that soil resuspension is responsible for the increased ^{137}Cs activity observed during the winter season. The mean ^{137}Cs (15 $Bq \cdot kg^{-1}$) and ^{40}K (300 $Bq \cdot kg^{-1}$) levels observed in soil around Bratislava (Daniel et al., 1996) 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).

What is specific for the Bratislava ^{137}Cs record when compared with similar results obtained for other localities (e.g., for Monaco as reported by Pham et al., 2005) is the existence of regular and wide winter maxima (Fig. 2) which may indicate a predominance of soil resuspension effects coupled with specific meteorological conditions.

A few single ^{137}Cs peaks observed usually during one month (e.g., in June 2005 and April 2006, Fig. 2) may indicate contributions from biomass burning of highly contaminated areas in Eastern Europe (Povinec et al., 2011), as

Saharan dust events had different time occurrence (Pham et al., in preparation). We cannot exclude, however, short-term increases in atmospheric ^{137}Cs levels due to transport of soil dust by strong winds when deeper soil layers are exposed to wind action. More detailed studies are therefore required when radionuclide data from short-term aerosol sampling is confronted with actual meteorological situation.

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 2007 was 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 constant.

(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 between the ^{137}Cs and ^{40}K atmospheric levels, $R = 0.84$). 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), which would also indicate a predominance of soil resuspension processes over the atmospheric concentrations of these radionuclides during the winter season.

Acknowledgements. This study was partially supported by the Scientific Grant Agency VEGA (project No. 1/0678/09), the Grant Agency APVV (project No. 0420-10), and by the EU Structural Funds (EU Research and Development Operational Programme funded by the ERDF, project No. 26240220004).

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Received on August 3, 2011.

Редактор *Е. И. Кравченко*

Подписано в печать 09.11.2011.
Формат 60 × 90/16. Бумага офсетная. Печать офсетная.
Усл. печ. л. 0,81. Уч.-изд. л. 1,13. Тираж 250 экз. Заказ № 57483.

Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
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