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# Migration of radiocaesium in individual parts of the environment

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**Abstract** The aim of this study was to compare the amount of radiocaesium in chosen compartments of environment in two localities of Košice vicinity (Košice and Jasov). Prevailing part of radiocaesium is in the upper layer of soils and specific activity of $^{137}Cs$ in the first layer for locality Jasov in 2001 achieved 21.49 Bq·kg$^{-1}$. Transfer factor values estimated for mixed grasses and for individual years varied within the range of 0.22 to 0.56 at locality Košice and of 0.09 to 0.19 at locality Jasov. On the base of results from modified Tessier sequential extraction method we determined that more than 50% of this radionuclide is in the soil in not extractable fraction. From studied species of mushrooms in sample of *Rozites caperata* the specific activity achieved 1822.0 Bq·kg$^{-1}$ d.w.

**Key words** $^{137}Cs$ • soil contamination • sequential extraction • mushrooms

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At present an increased attention is devoted to the occurrence of radionuclides in environment, due to the development of power supply, industry, agriculture, traffic, etc. The need of radionuclides study raised from several reasons. After penetration of these pollutants into environment arise their migration in the individual compartments of environment, their adsorption by natural matrices as well as intrusion into food chains. The existence of these contaminants in organisms can evoke various somatic and genetic changes and therefore decontamination of polluted areas is one of the most important subject of present research.

Monitoring of environment radioactivity is necessary because of determination of radioactivity influence on the health of today's and for future population. Our laboratory is one of the permanent component of radioactivity monitoring network in Slovak Republic, that carries out regular measurements of individual parts of environment, individual parts of food chains and working place.

Caesium belongs to the alkali metals and is the least inert and thus most reactive element in this group. Its chemical and metabolic-physiological reactions are similar to those potassium which latter is essential for many organisms and is enriched intracellularly. Radiocaesium does not occur naturally on earth, it is exclusively anthropogenic in origin. Radioisotopes of caesium are of environmental concern due to their relatively long half-life, emission of gamma radiation during decay and rapid incorporation into living organisms [11].

**Table 1.** Individual extraction agents and corresponding caesium fractions isolated from soil components

| Step | Reagent composition | Action time [h] | Isolated fraction |
|---|---|---|---|
| 1 | redistilled water (pH = 5.5) | 1 | water-soluble |
| 2 | 1 M $MgCl_2$ (pH = 7) | 1 | exchangeable |
| 3 | 0.025 M $Na_4P_2O_7$ | 1 | bound to humic acids |
| 4 | 1 M NaOAc + HOAc (pH = 5) | 24 | bound to carbonates |
| 5 | 0.04 M $NH_2OH{\cdot}HCl$ | 24 | bound to Fe/Mn |
| 6 | 30% $H_2O_2$ + $HNO_3$ (pH = 2) | 24 | organically bound and bound to sulfates |
| 7 | 2 M $NHO_3$ | 24 | residue, soluble in mineral acid |
| 8 | 1 M NaOH | 24 | residue, soluble in hydroxide |
| 9 | | | insoluble rest |

HOAc=$CH_3COOH$

The aim of our study was to determine the amount of caesium in chosen compartments of environment for two localities from Košice vicinity, i.e. Košice and Jasov.

Samples from Košice were collected in the area of Regional Public Health Authority, where our laboratory is situated. Jasov lies 28 km to the west of Košice. Since 1973 the greater part of this forest economic complex belongs to protected regional area called Slovenský kras. It is the greatest and the most typical area with the existence of the plain karst in the middle of the Europe. Therefore in 1977 United Nations Educational Scientific and Cultural Organisation declared it as a biospheric nature reserve.

## Materials and methods

Soil and plant samples were collected in the town Košice ($\lambda$ = 21° 1' E, $\varphi$ = 48° 43' N) and the vicinity of Košice–Jasov ($\lambda$ = 21° 57' E, $\varphi$ = 48° 41' N). Soil samples were taken from vertical profile 0–5, 5–15 and 15–30 cm. The soils under mushrooms were sampled from 0–10 cm. The lumps were crushed, dried separated from plant roots, homogenised and sieved to pass through 2-mm sieve.

Plants were harvested by cutting 5 cm above ground level and weight. Samples were dried at 105°C until total dehydration, homogenised and placed to Marinelli pots. For the studied plants $^{137}$Cs uptake was expressed as transfer factor. The soil-to-plant transfer factor is defined as: TF = activity concentration in plant (Bq·kg$^{-1}$ d.w.)/ activity concentration in soil (Bq·kg$^{-1}$ d.w.).

The fruiting bodies of mushrooms species (*Macrolepiota procera, Lepista personata, Lactarius piperatus, Lycoperdon perlatum, Clitocybe geotropa* and *Agaricus sylvaticus*) were collected in the locality Jasov in 2001. The other species were sampled (*Lactarius deliciosus*, *Lycoperdon perlatum*, *Russula mustelina*, *Armillaria mellea*, *Rozites caperata* and *Amanita phalloides*) in 2004.

For determination of individual fractions of radiocaesium in soils the modified Tessier sequential extraction method [10] was used where two steps were added to the original method [13]: extraction with redistilled water (step 1), and extraction with 2 M $HNO_3$ (step 7). Individual extraction agents and corresponding caesium fractions isolated from soil components are shown in Table 1. This experiment involved 125 g of dry soil and 500 ml of extraction agent placed into a 1000-ml bottle and shaken using end-over end shaker.

The specific activity of $^{137}$Cs in the studied samples was measured gamma-spectrometrically using multichannel analyser with Ge(Li) detector (Canberra Series 35 Plus). The data acquisition and analysis were performed using Gamat software.

## Results and discussion

The soil of these localities is characterised as brown earth with acidic soil reaction and high sorption capacity. Gamma-spectrometric analysis of individual soil layers showed that the specific activity of radiocaesium in the first layer for locality Jasov ranged from 15.4 to 21.49 Bq·kg$^{-1}$. Radiocaesium activity is gradually decreasing and in the third layer is lower by one third in comparison with first layer. Behaviour of the radiocaesium vertical distribution is similar in individual years for observed period (Fig. 1). Significantly lower values of radiocaesium were obtained for samples from locality Košice. For observed period values in the first

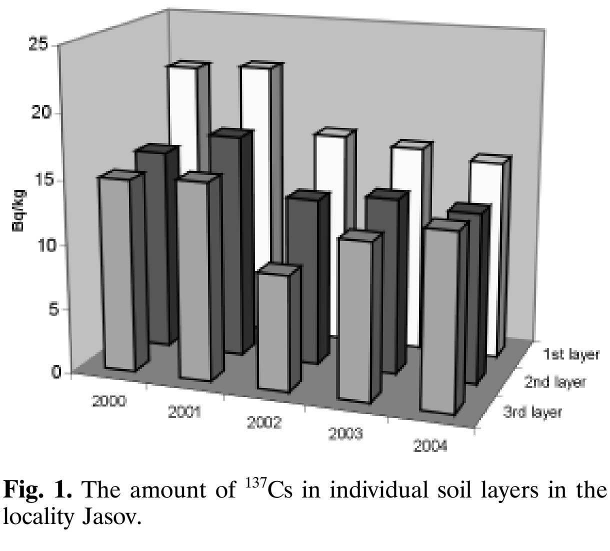


**Fig. 1.** The amount of $^{137}$Cs in individual soil layers in the locality Jasov.

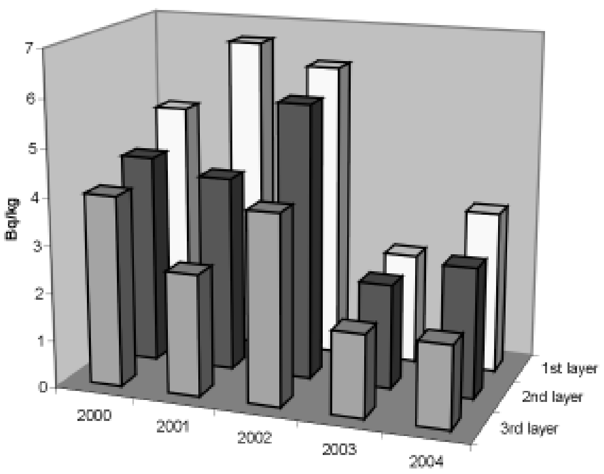


**Fig. 2.** The amount of $^{137}$Cs in individual soil layers in the locality Košice.

layer varied in the range from 2.32 to 6.63 Bq·kg$^{-1}$ (Fig. 2).

Radiocaesium migrates vertically in soils very slowly and prevailing part of caesium is in the upper layer of soil in dependence on soil type.

Samples of the soils under mushrooms were taken upon the 10 cm. This is because all forest herbs and most trees take up nutrients from the uppermost 10 cm of soil. Considerably higher values were determined in the forest soil that was sampled in 2001 ($^{137}$Cs = 182.8 ± 2.35 Bq·kg$^{-1}$ and $^{40}$K = 853.5 ± 20.1 Bq·kg$^{-1}$) and lower values were obtained in sample for year 2004 ($^{137}$Cs = 56.34 ± 1.40 Bq·kg$^{-1}$ and $^{40}$K = 565.30 ± 16.9 Bq·kg$^{-1}$).

It can be clearly seen that the main accumulation of caesium in the upper 10 cm of soil. Absorption of radiocaesium in soil is increasing with increased amount of organic matter. The high radiocaesium activity in the top layer is probably also due to the subsequent supply of radionuclides through dropped needles and leaching from needles and bark [7].

The surface of the area under study was randomly covered with grasses. We found that grasses covering controlled area accumulated amount of caesium. Values for two studied localities, i.e. Košice and Jasov are from the range of 0.28–0.58 Bq·kg$^{-1}$ f.w. and 0.34–0.79 Bq·kg$^{-1}$ f.w., respectively. Soil to plant relationship is evaluated by means of the transfer factor, which is usually expressed as TF (Bq·kg$^{-1}$ crop, d.w.)/(Bq·kg$^{-1}$ soil, d.w.). The soil activity concentration to a depth of 10 cm was used in the calculation. Transfer factor values estimated for mixed grasses and for individual years varied within the range of 0.22 to 0.56 at locality Košice and of 0.09 to 0.19 at locality Jasov (Fig. 3).

Experimental observations suggest that $K^+$ strongly suppress $Cs^+$ uptake [3, 15]. Plant roots absorb caesium less efficiently than its nutrient analogue, potassium. This is illustrated by the so-called Cs/K discrimination factor. This is defined as [(Bq·kg$^{-1}$ Cs in crop)/(Bq·kg$^{-1}$ K in crop)]/[(Bq·kg$^{-1}$ Cs in soil)/(Bq·kg$^{-1}$ K in soil)]. Discrimination factor for mixed samples of grass in localities Košice and Jasov ranged 0.15–0.34 and 0.08–0.15, respectively. DF values below unity indicate that K is

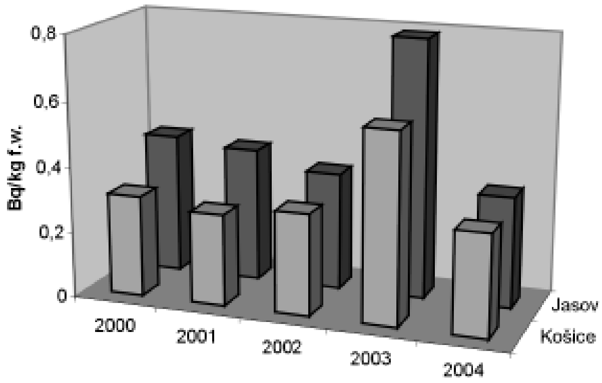


**Fig. 3.** The amount of $^{137}$Cs in grass samples in the localities Jasov and Košice.

more efficiently adsorbed than Cs. This fact is documented by lower transfer factors of mixed grass from locality Jasov, where are higher amount of potassium in soil in comparison with locality Košice.

Knowledge of the total content of radionuclides in soils provides a limited information only, and therefore, some measure of availability and mobility is required if reliable evaluations of pollution hazards are to be made [4, 6]. In order to stop radionuclides transfer into plants and its intrusion into food chains and groundwater it is necessary to know the extent and conditions of its binding to soil particles [1, 9, 14].

Our data presented in Fig. 4 shows very low caesium reactability from the given soil samples obtained by modified Tessier sequential extraction method. Water-soluble and exchangeable fractions that define intensity of vertical migration in soil samples is for caesium very low. For soil samples collected in 2001 and 2004 in the locality Jasov values are a little higher than 5% of total activity. In comparison with other fractions, the fraction soluble in mineral acid (in studied soil sample is about 11% of total activity) and fraction bound to organic matter are important, too. Prevailing part of this radionuclide is in the not extractable fraction as we

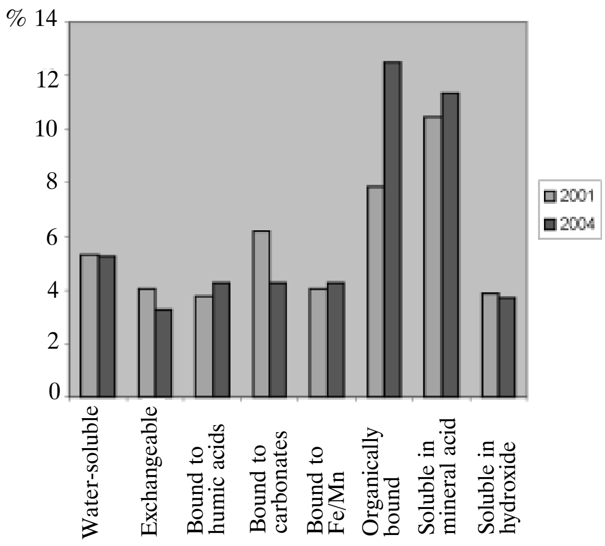


**Fig. 4.** The $^{137}$Cs individual fractions shares in the soil samples from the locality Jasov.

**Table 2.** The amount of $^{137}$Cs and $^{40}$K in the mushrooms from locality Jasov sampled in 2001

| Family | Species | [Bq·kg$^{-1}$ d.w.] | | [Bq·kg$^{-1}$ f.w.] | |
|---|---|---|---|---|---|
| | | $^{137}$Cs | $^{40}$K | $^{137}$Cs | $^{40}$K |
| *Lycoperdaceae* | *Lycoperdon perlatum* | 14.79 | 921.20 | 1.91 | 118.90 |
| *Tricholomataceae* | *Lepista personata* | 17.53 | 1843.00 | 1.17 | 123.20 |
| | *Clitocybe geotropa* | 9.79 | 1098.00 | 0.99 | 111.30 |
| *Russulaceae* | *Lactarius piperatus* | 4.46 | 1483.50 | 0.50 | 166.77 |
| *Agaricacea* | *Macrolepiota procera* – caps | 4.09 | 1695.00 | 0.31 | 128.08 |
| | *Macrolepiota procera* – stipes | 4.86 | 1280.00 | 0.37 | 98.18 |
| | *Agaricus sylvaticus* | 4.10 | 2017.00 | 0.31 | 151.60 |

assumed because of its strong affinity to clay minerals. Subdivision of radiocaesium in soil samples from individual collections in the locality Jasov is similar (Fig. 4). It can be concluded that extremely low concentration of caesium in soil solution is the factor limiting caesium uptake by the root systems [11].

Mushrooms are characterised by high ability to accumulate radiocaesium [2, 5, 7, 8, 12]. The reason why mushrooms work as such a good indicators of radioactivity in general is connected with their structure. Their bodies consist of gentle fibres, hyphae. Fungal metabolism differs from that of green plants. Mushrooms are heterotrophic organisms and depend on supply of organic compounds. Water constitutes about 90–95% of mushroom fresh weight.

The specific activity of examined species of mushrooms from locality Jasov collected in 2001 varied from 4.09 Bq·kg$^{-1}$ d.w. up to 17.53 Bq·kg$^{-1}$ d.w. (Table 2). Higher values of radiocaesium were obtained for the samples from the year 2004 (Table 3). Minimal activity was determined for *Lycoperdon perlatum*, i.e. 31.14 Bq·kg$^{-1}$ d.w. and in *Rozites caperata* the specific activity achieved 1822.0 Bq·kg$^{-1}$ d.w. According to literature [7] this species is characterised by the high ability to accumulate caesium. This fact can be confirm by a high value of discrimination factor that is equal 8.84. Interesting is a high value of $^{137}$Cs amount in *Amanita phalloides* as well as high discrimination factor, that is equal 2.59.

The fruiting bodies of three species *Armillaria mellea*, *Lactarius turpis* and *Clitocybe nebularis* in the locality Košice were collected. The data presented in Table 4 shows that the highest value of radiocaesium was determined for *Armillaria mellea*, although this one is about three times lower in comparison with the sample form Jasov. $^{137}$Cs is weakly cummulated by *Clitocybe nebularis*. This is an edible mushrooms but with low grade taste.

## Conclussions

On the base of obtained results we can conclude that radiocaesium migrates vertically in soils very slowly and prevailing part of caesium is in the upper layer. Uptake of radiocaesium by plants is suppressed by amount of potassium in soil. Extremely low concentration of caesium in soil solution is the factor limiting caesium uptake by the root systems, too.

**Table 3.** The amount of $^{137}$Cs and $^{40}$K in the mushrooms from locality Jasov sampled in 2004

| Family | Species | [Bq·kg$^{-1}$ d.w.] | | [Bq·kg$^{-1}$ f.w.] | |
|---|---|---|---|---|---|
| | | $^{137}$Cs | $^{40}$K | $^{137}$Cs | $^{40}$K |
| *Lycoperdaceae* | *Lycoperdon perlatum* | 31.14 | 1173 | 2.97 | 111.9 |
| *Tricholomataceae* | *Armilaria mellea* | 166.5 | 1699 | 17.86 | 182.3 |
| *Amanitaceae* | *Amanita phalloides* | 764.4 | 2954 | 33.37 | 129.0 |
| *Russulaceae* | *Lactarius delicious* | 50.46 | 1820 | 4.59 | 178.6 |
| | *Russula mustelina* – caps | 118.4 | 2036 | 7.83 | 134.6 |
| | *Russula mustelina* – stipes | 94.55 | 1996 | 8.75 | 184.7 |
| *Cortinariaceae* | *Rozites caperata* | 1822.00 | 2069 | 130.60 | 148.2 |

**Table 4.** The amount of $^{137}$Cs and $^{40}$K in the mushrooms from locality Košice

| Family | Species | [Bq·kg$^{-1}$ d.w.] | | [Bq·kg$^{-1}$ f.w.] | |
|---|---|---|---|---|---|
| | | $^{137}$Cs | $^{40}$K | $^{137}$Cs | $^{40}$K |
| *Tricholomataceae* | *Armilaria mellea* – 2002 | 68.77 | 2444.00 | 4.47 | 158.8 |
| | *Clitocybe nebularis* – 2004 | 20.24 | 2360.00 | 1.49 | 173.3 |
| *Russulaceae* | *Lactarius turpis* – 2004 | 4.39 | 2622.00 | 0.22 | 130.8 |

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