

COULD MUSHROOMS ACT AS CONCENTRATION PATHWAYS TO INTERNAL CONTAMINATION OF SOME MARGINALIZED COMMUNITIES BY ^{137}Cs IN SLOVAKIA?

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ABSTRACT

The aim of this study was to obtain some preliminary data necessary for the assessment of the possible existence of small-area local sources of contaminated mushrooms in forested areas with comparatively low inventory of man-made ^{137}Cs . On the study plot near Čierne-Svrčinovec (North-western Slovakia) 31 samples were collected during periodic sampling in the years 2012-2013 and analysed by gamma-spectrometry. Activities ^{137}Cs in mushrooms from entire sampling plot followed the lognormal distribution with extremely high variation and high positive skew, with projected three-sigma (99.75%) range from 0.49 Bq.kg⁻¹ to 1537 Bq.kg⁻¹ (dry weight, d.w.) and with geometrical average of 27.4 Bq.kg⁻¹ (d.w.). Another 11 samples were collected or bought from roadside vendors living in Roma slums near villages Richnava and Kluknava (Spiš region, eastern Slovakia). Obtained edible wild mushrooms were in a good shape despite prolonged drought indicating existence of well hidden forested sites in slum surroundings. Highest activity of ^{137}Cs in this set was recorded in *Boletus edulis* (263.3 ± 20.3) Bq.kg⁻¹ (d.w.). Both geometric mean (49.0 Bq.kg⁻¹) and expectation value (91.2 Bq.kg⁻¹ d.w.) of lognormal distribution were higher than that of the study plot on NW Slovakia. No sample exceeded the legal radiohygienic limit for radiocaesium contamination. The limited extent of presented study only slightly indicates the possibility of slow changes in spatial redistribution of bioavailable ^{137}Cs in forest ecosystem. However, available data on this subject from different sources suggest that there is no reason for massive occurrence of highly contaminated fruiting bodies in small areas surrounding settlements, where some critical groups of individuals, who are used to consume large amounts of mushrooms from limited geographical area could in such case develop significant internal contamination.

Keywords: ^{137}Cs , mushrooms, contamination, marginalized communities

INTRODUCTION

Nearly three decades after the Chernobyl accident and subsequent contamination almost of the entire European territory by a complex mixture of radionuclides there is still significant amount of ^{137}Cs activity present in various biological parts of the terrestrial ecosystems (Aarkrog, 1988; Gudiksen *et al.*, 1989; Bossew *et al.*, 2001; Lehto *et al.*, 2013). This is due to a large amount of ^{137}Cs released, it's long half-life ($T_{1/2} = 30.1$ y) and chemical similarity of caesium to potassium. Spatial distribution of deposited contamination was uneven on the continental scale and also on a much smaller local scales (Lettner *et al.*, 2000; Shoigu and Bolshov, 2006; Winkelbauer *et al.*, 2012). Outside the close surroundings of the contamination source individuals have been threatened only by doses received from internal contamination, originating at present almost exclusively from consumption of contaminated food (EC 1998; Povinec *et al.*, 2012).

In response to accident the protection of the public was accomplished by food restriction criteria applied by establishing maximum permitted levels of contamination (COUNCIL REGULATION (EURATOM) No 3954/87; EC, 2009). While food restriction criteria adopted in EU clearly achieved the objective of providing appropriate protection of individuals with ordinary consumer habits, some critical groups of individuals, who habitually consume large amounts of certain foods from limited geographical area and/or local food sources could exist (EC, 1998).

The behaviour of radionuclides in forest ecosystems differ substantially from the agricultural ecosystems, which are more generally considered in radiation dose assessments (Calmon *et al.*, 2009). Radionuclides can be trapped and recycled with particularly high efficiency in forests, resulting in long residence times and the potential for enhanced internal exposures delivered to human population over timescales of decades to centuries (Shaw, 2007; Nimis, 1996). Fungal and microbiological activity is likely to contribute substantially to the long-term retention of radionuclides in organic layers of the forest soil. Most of the isotope ^{137}Cs , the main contaminant released from Chernobyl (USSR, now Ukraine) can still be found in organic horizons of forest soil, more than two decades after the accident (Dupré de Boulois *et al.*, 2008; Winkelbauer *et al.*, 2012). Mycorrhizal fungi do have the biochemical capabilities to mobilize nutrients from polymeric sources of carbon and therefore are not themselves dependent upon decomposer fungi for such activities (Steiner *et al.*, 2002). Radiocaesium is expected to be stored predominantly within fungal biomass below ground, since the biomass of the fruit bodies constitutes only a small percentage of the total fungal biomass. Nikolova *et al.* (1997) demonstrated that the ^{137}Cs activity concentrations in fruit bodies and the fungal part of *Suillus variegatus* were about the same magnitude. Fungus-mediated translocation of ^{137}Cs into fresh litter is supposed to be one reason for the persistence of radiocaesium in the organic horizons of forest soil. Fungi are also considered to be responsible for the horizontal spatial redistribution and local enrichment of ^{137}Cs in fungal clusters (Nikolova *et al.*, 1997; Rafferty *et al.*, 2000). In the underlying mineral horizons,

the immobilization of radiocaesium by clay minerals is supposed to outweigh the potential fungus-mediated upward transport. First pure mineral horizon therefore acts as a sink for radiocaesium (Rühm et al., 1996). In the long time-scale (several years to decades) lateral redistribution of water-soluble cesium could occur especially in very rugged forested terrain, where the main role is played by surface water, periodic dissolution of snow cover, the activity of animals, humans and different mushroom species itself - their life cycle and changes in mycorrhizal systems on the site (Steiner et al., 2002). This implies the unpredictability of picking highly contaminated mushrooms even in forested landscapes with moderate or low levels of ^{137}Cs inventory. Though the amount of wild mushrooms consumed in many countries of central and east Europe is higher than in the rest of continent (for instance statistical mean of 5.6 kg of fresh mushrooms per household in Czech Republic, Kalač (2010)), more than an order of magnitude higher consumption rates could be estimated in some individuals of marginalized communities living in poverty at regions with highest unemployment in Slovakia, who are often looking for subsistence as gatherers in local forested areas. The aim of this study was to obtain some preliminary data necessary for the assessment of the possible existence of small-area local sources of contaminated mushrooms in forested areas with comparatively low inventory of ^{137}Cs .

MATERIAL AND METHODS

Study area, sampling

Mushrooms were collected during two seasons in 2012-2013 north of the village Čierne-Svrčinovec at Kysuce region (western Slovakia). The sampling area was a rectangle 0,35 km wide and 1,26 km long, heading northeast from the hill Košariská (618 m above sea level). Far sides of the rectangle were covered by forest, middle of the sampling plot were meadows in a shallow valley, crossed by a small stream. Spruce dominated forest was sparsely inhabited also with *Abies alba*, *Fagus sylvatica*, *Acer pseudoplatanus* and *Pinus sylvestris*. Long-term average precipitations are between 900 – 1000 mm. Generally, mushroom gathering seasons in years 2012-2013 were in the whole Slovakia exceptionally poor. Year 2012 was very drought and in the year 2013 the precipitation was unevenly distributed to early spring and late autumn.

Another sets of mushroom samples were purchased from roadside vendors from Roma slum near villages Richnava and Kluknava (Spiš region, eastern Slovakia) in August 2012 and collected at the same day at the neighbouring forest around the hill Poráč (760 m above sea level).

Analytical procedure

Mushrooms after transport into laboratory were weighted, sliced and dried at 90°C. Dried mushrooms were homogenised and poured to standard Marinelli beakers for gamma spectrometric analysis. High purity germanium detectors (CANBERRA, USA) with relative efficiency of 25% and resolution (FWHM) better than 1.9 keV@1332 keV, connected to modular spectrometric system (ORTEC and CANBERA, USA) were used for spectra acquisition. Typical live times varied between 90 000 s – 250 000 s depending on observed activity of sample and it's mass for achieving reasonable accuracy and/or low limits of detection. Obtained data were processed using proprietary gamma spectroscopy analysis software Asap (NUCLEAR DATA, USA) and GammaVision, v.4.1. (ORTEC, USA).

Lognormal distribution testing and descriptive data analysis were programmed in MS Excel 2010 using its internal statistical functions (Markechová et al., 2011).

RESULTS AND DISCUSSION

Study area near Čierne-Svrčinovec

Typical range surveyed by free-time mushroom collectors is usually much smaller than 10 km², which is the grid area per 1 measuring point used on the official map of ^{137}Cs activity over Slovakia territory (Gluch et al., 2005). For obtaining information on the range and distribution of ^{137}Cs activity in mushrooms a small gathering plot (0,44 km²) was chosen in a moderately hilly country near Čierne –Svrčinovec. During periodic sampling (8 sweeps in 2012, 3 in 2013) of the plot overall 31 samples with sufficient amount of mushrooms for analyses were collected. Fruiting bodies of species spotted in inadequate amounts on the whole area of sampling plot during one day were combined to mixed samples. Concentration of ^{137}Cs activities (Bq.kg⁻¹ dry weight, d.w.) in

mushrooms collected on both forested parts of sampling plot ranged from (248.8 ± 19.1) Bq.kg⁻¹ in *Paxilus involutus* to (6.03 ± 1.03) Bq.kg⁻¹ in *Amanita muscaria*. Observed range of ^{137}Cs activities around 1.5 order of magnitude for the whole set of species found on a sampling plot is at the lower end of the range of variations reported in more heavily contaminated single forests in Europe (Mietelsky et al., 2010; Guillén and Baeza, 2014). Having taken into account also two samples of *Macrolepiota procera*, picked-up outside of the forested parts of the sampling plot, observed minimum to maximum range increased to 2.5 magnitude of order, which is still in the range of variations given in literature. Low levels of ^{137}Cs found in fruiting bodies of *Macrolepiota procera* are in compliance with observations from different regions (Bazala et al., 2005; Dvořák et al., 2006; Kalač, 2012; Miššik, 2013). This may be explained by non-mycorrhizal character of *Macrolepiota* sp. and also by a significant difference between organic horizons complexity and dynamics in forest and more sparse vertical soil profiles on forest/grass meadow boundaries, preferred by this species.

On the whole territory of Slovakia yearly ranges varied between (2,9 – 4,1) orders of magnitude and total variation of more than 5 orders of magnitude were found for ^{137}Cs content in mushrooms collected in years 2009 – 2012 as a part of monitoring programme of State Veterinary and Food Administration of Slovak Republic (Miššik, 2013). However, annual surveys were done on a much larger spatial scale, with density of sampling more than 5.10⁴ times lower than the presented data.

Closer inspection of data reveals that activities of man-made ^{137}Cs in mushrooms from entire sampling plot followed the lognormal distribution (Kolmogorov-Smirnov test, $\alpha = 0.05$), while activities of primordial ^{40}K were distributed normally. In each sample the concentration of ^{40}K activity was well above the level of ^{137}Cs contamination. Average concentration of ^{40}K activity in mushrooms was (1127 ± 389) Bq.kg⁻¹ (d.w.). Concentration of ^{137}Cs activity in fruiting bodies averaged at 27.4 Bq.kg⁻¹ (d.w.), descriptive statistical analysis of data with extremely high variation and high positive skew projected three-sigma (99.75%) range from 0.49 Bq.kg⁻¹ to 1537 Bq.kg⁻¹.

Table 1 Ranges of activity concentrations of ^{137}Cs and ^{40}K (Bq.kg⁻¹ d.w.) in mushrooms collected during 2012 – 2013 on sampling plot near Čierne – Svrčinovec

Species	n	Cs-137		K-40	
		Bq.kg ⁻¹ d.w.		Bq.kg ⁻¹ d.w.	
		max.	min.	max.	min.
<i>Paxilus involutus</i>	2	248.8	÷	149.7	1353 ÷ 1146
<i>Pseudoclitocybe cyathiformis</i>	1	142.3	±	14.6	1448 ± 30.5
<i>Lactarius deliciosus</i>	1	101.4	±	8.7	1075 ± 7.4
Mixture of mushrooms	7	100.6	÷	39.6	1181 ÷ 822
<i>Russula ochroleuca</i>	2	48.5	÷	22.1	908 ÷ 850
<i>Hygrophoropsis aurantiaca</i>	1	36.8	±	4.5	642 ± 17.3
<i>Volvariella hypophitys</i>	1	36.6	±	3.1	1313 ± 13.4
<i>Lactarius piperatus</i>	1	30.0	±	2.9	834 ± 12.3
<i>Amanita muscaria</i>	7	27.2	÷	6.0	997 ÷ 767
<i>Russula turci</i>	4	26.1	÷	14.0	828 ÷ 907
<i>Russula risigallina</i>	1	22.1	±	2.9	908 ± 11.4
<i>Russula olivacea</i>	1	16.6	±	2.1	795 ± 28.3
<i>Macrolepiota procera</i>	2	≤ LLD = 2.29	÷	≤ LLD = 0.76	1035 ÷ 947

Legend: LLD – lower limit of detection

* In the case of n=1, observed activity concentrations with two-sigma combined uncertainty are given.

Expectation value of ^{137}Cs contamination of fruiting bodies from this site reached 67.4 Bq.kg⁻¹ (DW). Highly asymmetric shape of log-normal distribution with a long tail to larger values leads to underestimation of the centre of the data set by the arithmetic mean for a limited number of samples. In the case the number of samples go to infinity (n → ∞) the arithmetic mean value is supposed to reproduce the expectation value of the log-normal distribution correctly (Semizhon et al., 2009). Similarly high ranges of variation were found on 10-

and also 100-times larger study areas and higher sampling density in Belgium (Gillet and Crout, 2000). Among main factors influencing the observed differences in contamination levels of mushrooms are the spatial distribution of the ¹³⁷Cs inventory, particularly in the layer surrounding the mycelium, the depth and extent of mycelium, degree of mixing of organic and inorganic soil layers (Kruyts et al., 2004).

Surroundings of Richnava and Kluknava

For obtaining information about possible sites with steady yields of prized mushroom species, known only to experienced mushroom gatherers and hidden to casual picklers, quality mushrooms were bought from roadside vendors from Roma slum in Spiš region in August, 2012. Although at the time of acquisition of samples one of the worst mushroom gathering seasons was taking place due to a lasting drought, obtained fruiting bodies were fresh and in a very good shape. They were presumably picked on a sun-protected wet site, probably in some deeper depression. Such places have also higher probability to act as a secondary deposition sites to ¹³⁷Cs redistribution due to rainwater or melting snow.

Table 2 Ranges of activity concentrations of ¹³⁷Cs and ⁴⁰K in mushrooms bought or collected near villages Richnava and Kluknava (Spiš region, Eastern Slovakia) in August 2012

Species	n	Cs-137		K-40	
		Bq.kg ⁻¹ d.w.		Bq.kg ⁻¹ d.w.	
		max.	min.	max.	min.
<i>Boletus edulis</i>	4	263.3	÷ 63.7	745	÷ 428
<i>Sarcodon imbricatus</i>	1	69.5 ± 8.0		711 ± 87	
<i>Leccinum albostipitatum</i>	4	43.2	÷ 17.2	37	÷ 630
<i>Boletus reticulatus</i>	1	16.7 ± 2.4		820 ± 94	
<i>Neoboletus luridiformis</i>	1	11.5 ± 1.9		809 ± 94	

Legend: * In the case of n=1, observed activity concentrations with two-sigma combined uncertainty are given.

Concentration of ¹³⁷Cs activities (Bq.kg⁻¹ dry weight, d.w.) in mushrooms obtained from presumably neighbouring forested parts of both settlements ranged from (263.3 ± 20.3) Bq.kg⁻¹ in *Boletus edulis* to (11.5 ± 1.9) Bq.kg⁻¹ in *Neoboletus luridiformis*. Observed range of ¹³⁷Cs activities slightly above 1 order of magnitude for the whole set of samples is rather narrow, which could be caused not only by a very low number of samples, but particularly by a visible grouping of two purchased sets of mushrooms (*B. edulis* and *L. albostipitatum*) which suggests that they originated from a small sampling site. As in previous set of samples in each sample the concentration of ⁴⁰K activity was above the level of ¹³⁷Cs activity, but average concentration of ⁴⁰K activity (701 ± 210) Bq.kg⁻¹ (d.w.) was lower than the previous one. Geometrical average of ¹³⁷Cs activity concentration in fruiting bodies was 49.0 Bq.kg⁻¹ (d.w.), statistical analysis of lognormally distributed data produced narrower three-sigma (99.75%) range from 1.73 Bq.kg⁻¹ to 1386 Bq.kg⁻¹ with higher expectation value (91.2 Bq.kg⁻¹ d.w.) of ¹³⁷Cs contamination of mushrooms from this area. The fact that in the middle of a prolonged drought and very poor mushroom gathering season local prospectors were able to offer highly prized mushrooms in very good quality and quantity indicates that in the surroundings of the settlements there should occur suitable sites maintaining good conditions for fungal growth, in particular sufficient moisture. Despite the low average contamination of forest in this area, indicated on large scale radiological survey the analysis results also suggest that the same small sites could also act as catchment areas for the gradual accumulation of ¹³⁷Cs through lateral transport of soluble cesium from upper organic horizons of upstream slopes of the forest. Given the limited scope of this study, these arguments are poorly founded, but at least they could draw attention to the need of monitoring the situation in the forest ecosystem where cesium-137 can keep moving in closed cycles yet more than two centuries to the future at suitably shaped forested landscape formations on a small scale. Such sites could create favourable conditions for growth of wild edible mushrooms with higher levels of contamination by ¹³⁷Cs. However, data on observed levels of ¹³⁷Cs activity in mushrooms from Slovakia (Bazala et al., 2005; Dvořák et al., 2006; Kalač, 2012; Miššik, 2013) do not indicate a possibility of more than a rare

incidence of radiocesium contaminated mushrooms exceeding the radiohygienic limit of 600 Bq.kg⁻¹ fresh weight (EC, 2009). Only massive occurrence of such fruiting bodies in small areas around settlements could lead to increased risk from internal contamination in some critical groups of individuals, which are used to consume large amounts of mushrooms from limited geographical area (EC, 1998).

CONCLUSION

The limited extent of presented study only slightly indicates the possibility of slow changes in spatial redistribution of bioavailable ¹³⁷Cs in forest ecosystem. However, available data on this subject from different sources suggest that there is no reason for massive occurrence of highly contaminated fruiting bodies in small areas around settlements, where some critical groups of individuals, who are used to consume large amounts of mushrooms from limited geographical area could in such case develop significant internal contamination.

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