137Cs ACTIVITY CONCENTRATION IN MUSHROOMS FROM THE BOBRŮVKA RIVER VALLEY

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ABSTRACT
A total of 505 mushrooms belonging to 9 species were collected in the 2017 and 2018 mushrooming seasons near Dolní Rožínka in the Bohemian-Moravian Highlands and analysed by gamma spectrometry for 137Cs activity. The greatest 137Cs activity of 575 Bq kg\(^{-1}\) was detected in the species Boletus edulis, which is just below the permitted limit in its native state. In contrast, the detected activity level was only 316 Bq kg\(^{-1}\) in the mushroom Imleria badia, which is just below the permitted limit in its native state. However, differences in mean contamination values were not significant (p <0.05) due to high variability. It was shown that activity concentration is not dependent on the weight (size) of Imleria badia. Our results also confirmed the generally well known lower 137Cs activity in Russula species belonging to the group of gill-bearing or lamella-bearing mushrooms.

Keywords: radiocaesium; mushrooms; river valley; Boletus; Imleria

INTRODUCTION
The 137Cs isotope is one of more than 20 radioactive isotopes of caesium. 137Cs isotopes are formed at relatively high concentrations, reaching as much as 6.3 %, during the fission (nuclear chain) reaction in nuclear reactors (Söderlund et al., 2011).

Its negative biological effect is due to the emission of beta particles with the subsequent conversion of 137Cs to metastable 137Ba, which emits gamma photons of an energy of 661.7 keV (Wasek et al., 2014).

Despite the fact that more time than the physical half-life of 137Cs (30.17 years) has elapsed since the Chernobyl nuclear power plant accident (26 April 1986), there are still geographical areas with long-term persistent contamination (Smith and Beresford, 2005; Beňová et al., 2016). There are species of mushrooms in coniferous forests with a great ability to accumulate radiocaesium. Mushrooms probably contribute to the long-term retention of radiocaesium in organic layers of forest soils, especially spruce stands (Steiner, Linkov and Yoshida, 2000). The radiocaesium activity concentration in mushrooms from different areas of the Czech and Slovak Republics in 2000–2004 was reported by Dvořák et al. (2006).

The highest 137Cs activity concentration, representing 6.263 Bq kg\(^{-1}\) of dry matter, was measured in Imleria badia (syn. Xerocomus badius) from the area called Staré Ransko (the Bohemian-Moravian Highlands in the Czech Republic). The results also show significantly lower levels of 137Cs activity in Slovakia as compared to the Czech Republic.

137Cs activity depends not only on the contamination of the environment, but also on the mushroom species (Kalač, 2001). The highest levels of radioactive caesium contamination are found in saprophytic mushrooms (Duff and Ramsey, 2008). The mycelium of the non-edible mushroom Lactarius turpis (formerly L. necator) in Ukraine (1996 – 1998) showed an average activity of 52.700 Bq kg\(^{-1}\) (Vinichuk and Johanson, 2003).

Most of the published papers are based on mushroom collection from a relatively large area (Chiaravalle et al., 2018). In such large areas there are a number of different factors that affect the final mushroom contamination (Kalač, 2001; Dvořák et al., 2006).

First and foremost, the deposition of 137Cs is unevenly distributed depending on the precipitation intensity, latitude and altitude of the area (Lehto, Vaaramaa and Leskinnen, 2013; Bulko et al., 2014). The contamination of Europe by post-Chernobyl radiocaesium shows a mosaic pattern with significant differences in relation to area activity (Nilsson, 2009).
Scientific hypothesis
The aim of this work is to compare the $^{137}$Cs contamination of various mushrooms in a small relatively homogeneous conifer forest ecosystem in a river valley.

MATERIAL AND METHODOLOGY
Mushrooms were collected at a defined location in the Bobrůvka River Valley (near Dolní Rožinka in the Bohemian-Moravian Highlands) in the 2017 and 2018 mushrooming seasons. A total of 505 mushrooms of 9 different species, both common and rare in this particular area, were collected (Table 1).

Native mushroom samples were homogenised. Activity determination was carried out in the geometry of a 450ml Marinelli vessel or a 200ml polyethylene vial. These two geometries corresponded to the quantities of mushrooms per sample.

Determination of $^{137}$Cs activity was performed by two gamma spectrometry systems using HPGe GC4018 (40% efficiency) and HPGe GC2020 (20% efficiency) germanium detectors, both with a resolution of 1.8 keV, verified by the Czech Metrology Institute. The measurement time was 18 hours.

The programs Genie 2000 (Canberra) and Gamwin (Nuwa Třebíč) were used for evaluation. The $^{137}$Cs activity in all measured samples was higher than the minimum detectable activities (MDA). In addition to the mass activity, the total combined standard uncertainty was calculated using the formula:

$$ u_a = \left( \frac{u_E^2 + u_p^2 + u_y^2 + u_s^2 + u_r^2 + u_t^2 + u_A^2 + u_M^2}{u^2} \right)^{1/2} $$

where the individual standard uncertainties are as follows: relative uncertainty for efficiency ($u_E$), relative uncertainty for peak area ($u_p$), relative uncertainty for yield ($u_y$), relative uncertainty for time coincidence summing ($u_s$), relative uncertainty for electronic stability ($u_r$), relative uncertainty for time ($u_t$), relative uncertainty for decay ($u_A$), relative uncertainty for self-absorption ($u_M$), relative uncertainty for reproducibility ($u_M$).

Statistical analysis
The weighted arithmetic mean, average total standard uncertainty, and maximal and minimal value of mass activity were calculated for statistical evaluation. A two-sample unequal variance t-test was used to compare the mean differences. A correlation coefficient was calculated to compare the dependencies.

RESULTS AND DISCUSSION
Mushrooms, as one of the most important components of the forest ecosystem, are able to accumulate a significant amount of radionuclides, including $^{137}$Cs (Heinrich, 1992; Škrlak et al., 2013; Guillen and Beaza, 2014). The first mention of contamination of mushrooms with radiocaesium comes from the nineteen sixties. Mass activity values in a range of 10 to 2.500 Bq kg$^{-1}$ have been published in Germany (Grüter, 1964 and 1971). According to Kalač (2001), contamination is often from nuclear weapons tests. Other factors that can affect the value of mushroom contamination in addition to the growth medium properties are the climate and the associated amount of precipitation, as well as the sampling time and the fungus type itself (Heinrich, 1993). Furthermore, it has been found that the levels of radiocaesium in the underground parts of fungi (mycelium) are higher than the $^{137}$Cs levels in the aboveground parts such as the medulla and the cap (Viníček and Johanson, 2003). The distribution of radiocaesium in various above-ground parts of fungi is uneven, with higher activity found in fungi caps (Heinrich, 1993; Mukhopadhyay et al., 2007). Mushroom samples collected in coniferous forests are characterised by a high content of radionuclides compared to mushroom samples collected in deciduous forests (Čípková, 2004). $^{137}$Cs activity ranged between 273 and 1.165 Bq kg$^{-1}$ in fresh mushrooms collected in the French Alps in 1999 – 2002 (Pourcelot et al., 2003). In contrast, the highest contamination measured in the dry matter of mushrooms originating in the Bohemian-Moravian Highlands (Czech Republic) was 2.263 Bq kg$^{-1}$. Contamination levels in Slovakia were significantly lower (Dvořák et al., 2006).

Research conducted in Europe, Japan and North America in 2008 has shown that Boletus subtomentosus is the species with the highest contamination levels of the entire Boletus family. The potential danger represented by this mushroom is even bigger due to its presence in large numbers in all types of forests (Duff and Ramsey, 2008). However, we could not find this mushroom species in the Bobrůvka River Valley.

The mushrooms analysed in our study were collected from a small area in pine tree habitats in areas of steeply sloping hillsides modelled by river erosion. The results for $^{137}$Cs mass activities are shown in Table 1. The largest $^{137}$Cs activity was found in the species Boletus edulis, amounting to 1.575 Bq kg$^{-1}$ in native state, which is just below the legal limit. However, this species also showed the greatest variation range, as mushrooms with a mass activity of 28 Bq kg$^{-1}$ were also found. In contrast, the greatest activity seen in Inlmeria badia, which as a representative of the Boletaceae family is often quoted in connection with the highest cumulative ability, was only 316 Bq kg$^{-1}$. The differences between the average contamination values of both mushroom species (197 and 173 Bq kg$^{-1}$) were not, however, significant due to high variability. Kunová, Dvořák and Beňová (2006) reported the highest $^{137}$Cs activity value measured in Inlmeria badia from the Staré Ransko locality (the Bohemian-Moravian Highlands). This activity reached 708 Bq kg$^{-1}$, which is around twice that of the mushrooms studied in our work. Another representative of the Boletaceae family is Xerocomellus chrysenteron. The variation range is from 19 to 2 Bq kg$^{-1}$, which shows a very low ability to accumulate radiocaesium. Given the 33 specimens monitored, this cannot be a coincidence. The results for Neoboletus luridiformis are also surprisingly low. In the 7 specimens found, the maximum concentration of contamination measured was 73 Bq kg$^{-1}$. 

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**Table 1**

| Mushroom Species | Mass Activity (Bq kg$^{-1}$) |
|------------------|-----------------------------|
| Boletus edulis    | 1.575                       |
| Inlmeria badia    | 316                         |
| Xerocomellus chrysenteron | 19 - 2             |
| Neoboletus luridiformis | 73              |
**Table 1** Mass activity [Bq kg\(^{-1}\)] \(^{137}\)Cs in different species of mushrooms.

| Mushroom species          | n (number) | Mean Bq kg\(^{-1}\) | CNS* Bq kg\(^{-1}\) | Max Bq kg\(^{-1}\) | Min Bq kg\(^{-1}\) |
|---------------------------|------------|---------------------|---------------------|---------------------|---------------------|
| *Imleria badia*           | 113        | 173                 | 9.7                 | 316                 | 11                  |
| *Boletus edulis*          | 19         | 197                 | 12.9                | 575                 | 28                  |
| *Neoboletus luridiformis* | 7          | 27                  | 2.7                 | 73                  | 10                  |
| *Xerocomellus chrysenteron* | 33       | 13                  | 1.5                 | 19                  | 2                   |
| *Suillus grevillei*       | 1          | 28                  | 7.4                 |                     |                     |
| *Russula aeruginosa*      | 29         | 27                  | 1.9                 | 39                  | 9                   |
| *Russula olivacea*        | 3          | 18                  | 2.4                 | 5                   | 24                  |
| *Russula vinosopurpurea*  | 2          | 13                  | 1.7                 | 5                   | 20                  |
| *Russula ochroleuca*      | 1          | 9                   | 1.3                 |                     |                     |

Note: * Average combined standard uncertainties \(u\_c\), (Guide to the Expression of Uncertainty in Measurement for Standardization).

Furthermore, the dependence between the average weight of the mushrooms and their activity was also studied. For *Imleria badia*, the highest activity level of 316 Bq kg\(^{-1}\) was measured in a mushroom sample weighing 25 g, while the lowest activity level of 56 Bq kg\(^{-1}\) was measured in a mushroom sample weighing 24 g. The mushroom sample with the lowest weight (8 g) had an activity level of 106 Bq kg\(^{-1}\).

**CONCLUSION**

The results show that in the monitored area *Boletus edulis* shows a higher capacity of \(^{137}\)Cs contamination as compared to the more frequently occurring *Imleria badia*. No correlation was found between the radiocaesium mass activity and the size of the collected mushrooms. No \(^{137}\)Cs activity above the limit was found in mushrooms in the monitored area in the years 2017 – 2018.

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