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^{137}Cs in mushrooms from Croatia sampled 15–30 years after Chernobyl

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ABSTRACT

The aim of this study was to select species with higher potential to accumulate ^{137}Cs among the available mushroom species, by determining the activity concentrations of ^{137}Cs in mushrooms collected along north and north-western part of Croatia. A total of 55 samples of 14 different species were analyzed and the potential of mycorrhizal and saprotrophic species to accumulate ^{137}Cs was compared. A wide range of the dry weight activity concentrations of ^{137}Cs was detected, ranging from 0.95 to 1210 Bq/kg (154 Bq/kg mean value; 52.3 Bq/kg geometric mean) in mycorrhizal and 1.05–36.8 Bq/kg (8.90 Bq/kg mean value; 5.49 Bq/kg geometric mean) in saprotrophic species. Statistical analyses showed that mycorrhizal species accumulate significantly higher concentrations of ^{137}Cs and thus could perform better as long-term bioindicators of environmental pollution by radiocaesium then saprotrophic species. The comparison of *Boletus* sp. and *Hydnum repandum* (both mycorrhizal species commonly found in Croatia) showed, in general order of magnitude, higher accumulation in *Hydnum repandum.* Clearly, mushrooms, especially mycorrhizal species, can be used as significant indicators even decades after the occurrence of any serious ^{137}Cs contamination event. However, as a wide range of values indicates that various parameters may influence the total uptake of the ^{137}Cs into the mushroom fruit bodies, it is necessary to emphasize that ^{137}Cs activity detected in a single mushroom sample is very site-specific.

1. Introduction

Monitoring of anthropogenic radionuclide pollution of the environment is of high importance, particularly if related to nuclear weapons testing and accidents at nuclear power-plant facilities. Search for the most appropriate bioindicating organisms in the specific geographic area and for a specific radionuclide has been carried out extensively, especially after the Chernobyl nuclear accident in 1986, with emphasis on radiocaesium. Along with ^{90}Sr, ^{137}Cs is the most significant long-lived radionuclide produced in those types of incidents. The accumulation of radiocaesium in different mushroom species is widely studied (Byrne, 1988; Kalač, 2001; Baeza et al., 2004; Dighton et al., 2008; Kioupi et al., 2015; Zalewska et al., 2016). The focus of most of the studies is radioactive concentration in mushroom fruit bodies along the specific geographic area and the dose estimation for the population due to their ingestion. A large portion of the pre-Chernobyl ^{137}Cs detected in fungi (Dighton and Horrill, 1988; Byrne, 1988; Giovani et al., 1990) indicates that fungi could be used as indicators of radiocaesium presence in the environment long time after the contamination. The aim of this work is to identify ^{137}Cs accumulators among the available mushroom species from the north and north-western part of Croatia, regardless of their edibility.

Generally, a wide range of ^{137}Cs concentrations in fungal fruit bodies was observed in most of the studies, not always following the level of contamination of the sampling sites (Olsen, 1994; Kioupi et al., 2015). Olsen reported more than 50 times higher (in some cases up to 100 times) radiocaesium concentrations in mycorrhizal fungal fruit bodies than in some other vascular plants sampled on the same site. It is obvious that the fungi differ in their potential to accumulate Cs (Clint et al., 1991). A thorough investigation of each transfer step in the process is essential (Clint and Dighton, 1992; Dupré de Boulois et al., 2008). Numerous factors influence ^{137}Cs uptake by mushrooms, such as distribution and density of fungal mycelia (Giovani et al., 1990; Kammerer et al., 1994), soil or substrate with its characteristics which also vary in depth (such as soil organic matter content and pH) (Guillitte et al., 1994; Yoshida and Muramatsu, 1994; Rühm et al., 1997), forest type and fruit body location (Grueter, 1964; Andolina and Guillitte, 1990; Vinichuk and Johanson, 2003) or trophic status (saprotrophic, parasitic or mycorrhizal). Often, a higher ^{137}Cs concentration is detected in mycorrhizal mushrooms, if compared to saprotrophic or parasitic ones (Guillitte et al., 1994; Kammerer et al., 1994; Yoshida and Muramatsu, 1994; Gillet and Crout, 2000). Saprotrophes grow on organic matter while parasitic mushrooms grow attached to other species in a non-symbiotic relationship. Mycorrhizal species, on the

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other hand, form a close symbiotic relationship with their host plant through underground mycorrhiza. Therefore, it is reasonable to expect for them to retrieve more cesium via their mycorrhizal association with plant roots. Various species, both saprotrophic (S) and mycorrhizal (all of them ectomycorrhizal) (M), collected during the years 2012 and 2016 from differently contaminated parts of north and north-western Croatia, were analyzed by gamma-spectrometry to determine the ^{137}Cs mass activity concentrations. In order to obtain information about the contamination level of the sampling sites, soil samples from the same areas were collected and analyzed.

Due to a large range of ^{137}Cs activities found in mushrooms within the same group, especially mycorrhizal, the comparison between two very common ectomycorrhizal fungi – *Boletus* sp. and *Hydnum repandum* was made. *Boletus* mushrooms express high affinity for Cs (Heinrich, 1993; Kammerer et al., 1994; Toal et al., 2002; Vinichuk and Johanson, 2003; Řanda and Kučera, 2004), while some studies declare *Hydnum repandum* as one of the greatest Cs accumulators among mushrooms (Heinrich, 1993; Kammerer et al., 1994; García et al., 2015). As emphasized in previous studies (Mietelski et al., 1994; Shutov et al., 1996; Skuterud et al., 1997; Kalač, 2001), it should be indicated that in case of elevated ^{137}Cs activity in mushrooms, the annual dose due to their consumption should not be neglected.

2. Materials and methods

2.1. Mushrooms and soils sampling and sample preparation

A total of 55 samples of 14 different mushroom species were collected during 2012 and 2016. In 2012, the samples were collected, mainly randomly, from the three broader areas: 1) North-western Croatia (NWC) covering the area of about 4000 km^2, 2) Gorski Kotar area including surrounding areas and a part of Istria (GK) covering the space of about 4000 km^2 and 3) Banovina (B) covering the space of about 1000 km^2. Additional samples were collected in 2016, from the same locations - Banovina area and a smaller part of Gorski Kotar in vicinity of Vrbovsko (GKV) which covers an area of about 10 km^2. Both saprotrophic and mycorrhizal mushrooms were collected. The largest number of mycorrhizal species belonged to the genus *Boletus* (10 of *B. aestivalis* (Paulet) Fr., 8 *B. edulis* Bull., 2 *B. erythropus* Pers.) and a small number to other species – 2 samples of *Tricholoma portentosum* (Fr.) Quel., 3 *Lactarius* (1 *L. deterrimus* Groger, 1 *L. piperatus* (L.) Pers. and 1 *L. vellereus* (Fr.) Fr.), 2 *Leccinum griseum* (Quel.) Singer samples, 1 sample of *Russula cyanoxantha* (Schaeff.) Fr. and 1 of *Hydnum repandum* L. Saprotrophic species collected were as follows: *Agaricus campestris* L. (4), *Macrolepiota procera* (Scop.) Singer (7), *Clitocybe* (3 of *C. inversa* (Scop.) Quel., 4 of *C. nebularis* (Batsch) P.Kumm. species) and *Armillaria mellea* (Vahl) P. Kumm. (8). The number of individuals making one sample varied depending on their availability on each sampling site. The whole fruit bodies of mushrooms were collected and the samples were prepared from their stems and caps. The mushrooms were cleaned from any adhering debris of foreign material (plant, soil) by a plastic knife and dried at 50 °C until constant weight was reached, then ground and homogenized before being placed into cylindrical counting vessels of 125 cm^3. The mass of the dry mushroom samples packed into the given geometry ranged from 10 g up to 130 g.

Additionally, to compare two very common mycorrhizal fungi – *Boletus* sp. and *Hydnum repandum,* ^{137}Cs activity concentrations in fresh weight of mushrooms from Gorski Kotar area collected in 2001 and 2006 are included in the study. The stems and caps of these mushrooms were cleaned, cut and placed into the identical cylindrical counting vessels (125 cm^3). The mass of the fresh mushroom samples packed into the given geometry ranged from about 100 g up to 120 g.

Mushroom sampling sites were placed in vicinity of locations wherefrom soil samples in the NWC (39 samples) and GK areas (53 samples) were collected in the period 2000–2010. Considering that soil samples from these two areas were not collected along with mushrooms, the care was taken to collect mushrooms grown at same type of soils that were previously sampled in the vicinity. Soils from B (14 samples) and GKV (7 samples) areas were sampled along with mushrooms during the summer 2016. Each bulk soil sample was collected from the surface to the depth of 10 cm of open vertical soil profile. The soil was dried at 105 °C to a constant weight and homogenized before being placed into counting vessels of identical geometry (125 cm^3). The mass of the soil samples ranged between 100 and 180 g.

Table 1
^{137}Cs activity concentrations in dry weight mushroom tissue for the mushrooms collected in 2012 and 2016 (ref. date 1st of July 2016).

Species (Scientific name)	Location[a]	Year of collection	^{137}Cs activity (Bq/kg) dw
Mycorrhizal species (M)			
Boletus aestivalis	NWC	2012	33.2 ± 2.22
Boletus aestivalis	NWC	2012	15.9 ± 1.10
Boletus aestivalis	NWC	2012	16.3 ± 1.08
Boletus aestivalis	NWC	2012	64.8 ± 4.08
Boletus edulis	NWC	2012	22.9 ± 1.58
Boletus edulis	NWC	2012	41.9 ± 2.82
Tricholoma portentosum	NWC	2012	63.8 ± 3.97
Tricholoma portentosum	NWC	2012	48.8 ± 3.03
Lactarius deterrimus	NWC	2012	9.80 ± 0.75
Boletus aestivalis	GK	2012	1210 ± 73
Boletus aestivalis	GK	2012	240 ± 14.6
Boletus aestivalis	GK	2012	24.1 ± 1.80
Boletus aestivalis	GK	2012	12.8 ± 1.01
Boletus edulis	GK	2012	168 ± 10.3
Boletus edulis	GK	2012	170 ± 10.6
Boletus edulis	GK	2012	705 ± 42.6
Boletus edulis	GK	2012	308 ± 18.8
Boletus edulis	GK	2012	266 ± 16.1
Boletus edulis	GK	2012	79.9 ± 4.90
Boletus erythropus	GK	2012	355 ± 21.5
Boletus aestivalis	B	2012	27.7 ± 1.77
Boletus erythropus	B	2012	312 ± 19.0
Boletus aestivalis	B	2016	46.3 ± 2.92
Leccinum griseum	B	2016	52.2 ± 3.29
Leccinum griseum	B	2016	8.66 ± 0.59
Lactarius piperatus	B	2016	0.95 ± 0.36
Russula cyanoxantha	B	2016	7.75 ± 0.53
Lactarius vellereus	GKV	2016	5.11 ± 0.39
Hydnum repandum	GKV	2016	151 ± 9.11
Saprotrophic species (S)			
Agaricus campestris	NWC	2012	3.10 ± 0.62
Agaricus campestris	NWC	2012	4.46 ± 0.49
Agaricus campestris	NWC	2012	1.05 ± 0.29
Macrolepiota procera	NWC	2012	4.11 ± 0.52
Macrolepiota procera	NWC	2012	1.74 ± 0.28
Macrolepiota procera	NWC	2012	11.6 ± 0.83
Clitocybe inversa	NWC	2012	2.22 ± 0.48
Clitocybe inversa	NWC	2012	5.21 ± 0.70
Clitocybe inversa	NWC	2012	14.6 ± 0.98
Clitocybe nebularis	NWC	2012	6.07 ± 0.62
Armillaria mellea	NWC	2012	3.39 ± 0.41
Armillaria mellea	NWC	2012	1.13 ± 0.32
Armillaria mellea	NWC	2012	10.9 ± 0.82
Armillaria mellea	NWC	2012	10.3 ± 0.76
Agaricus campestris	GK	2012	8.00 ± 1.12
Macrolepiota procera	GK	2012	30.6 ± 0.65
Macrolepiota procera	GK	2012	1.59 ± 0.44
Macrolepiota procera	GK	2012	5.91 ± 0.51
Clitocybe nebularis	GK	2012	36.8 ± 2.50
Clitocybe nebularis	GK	2012	30.3 ± 2.07
Armillaria mellea	GK	2012	2.76 ± 0.53
Armillaria mellea	GK	2012	5.77 ± 0.62
Macrolepiota procera	B	2012	2.10 ± 0.41
Armillaria mellea	B	2012	16.4 ± 1.12
Clitocybe nebularis	GKV	2016	2.57 ± 0.37
Armillaria mellea	GKV	2016	8.92 ± 0.76

[a] NWC – North-western Croatia; GK – expanded Gorski Kotar area; B – Banovina; GKV - Vrbovsko (in Gorski Kotar area).