

## RADIOACTIVITY OF EUROPEAN WILD GROWING Edible Mushrooms

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Chapter 10

## RADIOACTIVITY OF EUROPEAN WILD GROWING EDIBLE MUSHROOMS

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#### ABSTRACT

Wild growing mushrooms are widely consumed as a delicacy in several European countries, at level up to several kg per year per capita. Activity concentrations of the natural isotope  ${}^{40}$ K are usually 800-1,500 Bq kg<sup>-1</sup> dry matter (DM). Other natural radionuclides with leading  ${}^{210}$ Pb and  ${}^{210}$ Po are of lower importance. Activities of  ${}^{137}$ Cs from nuclear weapons testing below 1,000 Bq kg<sup>-1</sup> DM were commonly reported until 1986. The situation changed dramatically after the accident of Chernobyl nuclear power station in 1986. Activities up to over 100,000 Bq kg<sup>-1</sup> DM of <sup>137</sup>Cs and to a lesser extent of <sup>134</sup>Cs were observed in some edible species in the following years. Commonly, mycorrhizal species accumulate radiocesium more than species with saprotrophic or parasitic nutritional strategy. Xerocomus badius, X. chrysenteron, Suillus variegatus, Rozites caperata, Laccaria amethystina and Hydnum repandum belong among the radiocesium highly accumulating and widely consumed species. Activity concentrations have been affected by several environmental factors, such as rate of soil contamination by the Chernobyl fallout, the depth from which mycelium takes nutrients and time since the accident. Most of the <sup>137</sup>Cs in forest soils appear to be available for uptake by mushrooms until now. A considerable consumption of accumulating species collected from the sites heavily contaminated in 1986 can be still of a health concern. The contamination can be reduced by soaking or cooking of dried or frozen mushroom slices. Until now, meat of wild boars eating some mushroom species from heavily contaminated areas can highly surpass statutory limit for <sup>137</sup>Cs.

**Keywords:** edible mushrooms; radioactivity; radionuclides; radiocesium; health concern; effective dose; game meat.

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## **1. INTRODUCTION**

The collection of wild growing edible mushrooms (higher fungi, macrofungi) as a delicacy has historical tradition in various parts of the world. This recreational and sometimes economic activity has been widespread e.g. in Slavonic countries of central and east Europe. Thus, a possibility of its deprivation or limitation due to the fears of mushroom contamination is considered as an abatement of lifestyle. Such situation arose in years following disaster of Chernobyl nuclear power station in 1986.

Radionuclide contamination of the environment has originated from the fallout following nuclear weapons testing, operation of nuclear energy-generating industries including their accidents and from medical uses of radioisotopes. Fungi (both filamentous and fruit-forming) are very efficient in absorbing radionuclides and are an important component of long-term accumulation of radionuclides owing to the long-living and huge hyphal network and biomass in upper horizons of forest soils.

High radioactivity levels of some wild growing mushroom species were reported in the 1960's (Grüter, 1964). The contamination of most of Europe with radioactive fallout from the Chernobyl accident triggered the extensive research and monitoring of the environment including mushrooms. Numerous original papers on mushroom radioactivity were published during the last 25 years. Moreover, several reviews of the literature are available. They focus either on environmental aspects and transfer particularly of radiocesium to fungi (both micro-fungi and mushrooms) (Gillett and Crout, 2000; Duff and Ramsey, 2008; Dighton et al., 2008) or on aspects of radionuclides intake from the consumption of various mushroom species (Kalač, 2001).

This chapter summarizes recent information on the radioactivity of European edible mushroom species. Only the weightiest articles until 2000 will be cited, further papers from that period are referred in the above mentioned reviews.

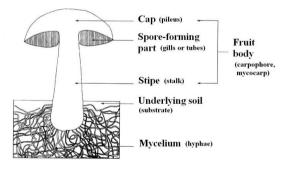


Figure 1. A sketch of a typical mushroom.

## 2. MYCOLOGICAL TERMS

Some mycological terms used in this chapter are given in Figure 1. The term mushroom will be used for the fruit body, mostly aboveground, of higher fungi. A fruit body is formed from spacious underground mycelium by the process of fructification. The lifetime of the bulk of fruit bodies is only 10-14 days.

According to their nutritional strategy, mushrooms can be divided into three groups. Mycorrhizal (symbiotic) species form a close, mutually favorable relationship with their host plant, mostly a tree. Saprotrophic species (or saprophytes) live on and metabolically consume organic matter. Parasitic species live on other species in a non-symbiotic relationship.

#### **3. RADIOACTIVITY UNITS AND LEGISLATION**

Published data on radioactivity levels are usually expressed per dry matter (DM) content. For unification, the commonly accepted content of 10% of dry matter in mushroom fruit bodies was used for the calculation in this chapter, if published data were given per fresh matter.

One Bq (becquerel) has been the unit for the activity of a radioactive source in which, on average, one atom decays per second. Activity concentration, i.e. activity per dry matter unit, will be used in this chapter. Within the European Union (EU), food statutory limits for radiocesium are 600 and 370 Bq per kg of fresh matter for adults and children, respectively. Thus, the limit for adults would be 6,000 Bq per kg of dry matter for mushrooms. Codex Alimentarius (FAO/WHO, 1991) defines explicitly the intervention levels for foodstuff being 1,000 Bq kg<sup>-1</sup> (fresh matter) for  $^{137}$ Cs.

Nevertheless, the considerable increase of foodstuff radioactivity following the Chernobyl disaster induced within the EU (formerly the EC) series of legislative provisions and recommendations (e.g. 2003/274/EC for mushrooms). Under the regulation from 1987, the maximum permitted level of <sup>137</sup>Cs was 1,250 Bq kg<sup>-1</sup> fresh matter (i.e. 12,500 Bq kg<sup>-1</sup> DM) for mushrooms. A similar limit of 1,000 Bq kg<sup>-1</sup> fresh matter (i.e. 10,000 Bq kg<sup>-1</sup> DM) was recommended by the International Atomic Energy Agency (IAEA) in 1994.

A possible risk of radioactivity for human health is expressed as the effective dose (E) given in mSv (millisievert) per year. The acceptable yearly burden for an adult, of the public, recommended by the IAEA (1996) has been 1 mSv. A contribution to the yearly effective dose to an adult from mushroom consumption can be calculated as follows:

 $\mathbf{E} = \mathbf{Y} \mathbf{x} \mathbf{Z} \mathbf{x} \mathbf{d}_{\mathbf{c}} ,$ 

where Y = annual intake of mushrooms (kg DM per person),

Z = activity concentration (Bq kg<sup>-1</sup> DM),

 $d_c$  = dose coefficient (conversion factor) defined as the dose received by an adult per unit intake of radioactivity. The values are 1.3 x 10<sup>-8</sup>, 1.9 x 10<sup>-8</sup>, 6.2 x 10<sup>-9</sup>, 6.9 x 10<sup>-7</sup> and 1.2 x10<sup>-6</sup> Sv per Bq for <sup>137</sup>Cs, <sup>134</sup>Cs, <sup>40</sup>K, <sup>210</sup>Pb and <sup>210</sup>Po, respectively.

## 4. NATURAL RADIONUCLIDES IN MUSHROOMS

Mushrooms contain considerably higher levels of potassium than foods of plant origin (see chapter 6 of this book). Potassium is not distributed evenly in a fruit body. The content usually decreases in order cap > stipe > gills or tubes in spore-forming part > spores. The accumulation factor (ratio of the contents in fruit body DM and in DM of underlying soil) is usually between 20 and 40. The radionuclide <sup>40</sup>K is present in the mixture of potassium isotopes at the constant level of 0.017%. Its half-life is extremely long.

Usual activity concentration of <sup>40</sup>K in numerous species of wild growing mushrooms, mostly from central Europe, were 800-1,500 Bq kg<sup>-1</sup> DM between 1984 and 1992 (Kalač, 2001 and references therein). Transfer factors for <sup>40</sup>K from underlying soil to fruit bodies exceeding value of 10 were reported for *Xerocomus badius*, *Amanita rubescens* and *Lycoperdon perlatum* (Eckl et al., 1986).

Data on <sup>40</sup>K published during the last decade are given in Table 1. The values are very similar to those reported until 2000. Thus, it seems that the incorporation of the stable potassium isotopes <sup>39</sup>K and <sup>41</sup>K and hence also <sup>40</sup>K is self-regulated by physiological requirements of a mushroom. The extremely high activity concentration of <sup>40</sup>K up to 12,000 Bq kg<sup>-1</sup> DM was reported in *Laccaria laccata* (Mietelski et al., 2010).

Unlike <sup>137</sup>Cs from radioactive fallout, natural <sup>40</sup>K is distributed in vertical profile of forest soils evenly (Krolak et al., 2010).

Isotope <sup>210</sup>Pb (half-life 22 years), a decay product of naturally occurring <sup>238</sup>U, <sup>226</sup>Ra or <sup>222</sup>Rn, is present in mushroom fruit bodies in activity concentration usually for two orders of magnitude lower than <sup>40</sup>K (Table 2). Activity concentrations of <sup>210</sup>Pb are consistently higher – often by one order of magnitude – than those of its long-lived precursor <sup>226</sup>Ra (see below). This indicates that a very small <sup>210</sup>Pb proportion, if any, originates from the decay of <sup>226</sup>Ra taken up from the soil (Kirchner and Daillant, 1998). Dose coefficient of <sup>210</sup>Pb is over 100 times higher than that of <sup>40</sup>K (see the previous section) and the effective dose from its intake from mushrooms should not be overlooked. For instance, such situation was observed in northern Finland with low contamination with the Chernobyl radiocesium (Vaaramaa et al., 2009).

A further natural radionuclide of <sup>238</sup>U decay, <sup>210</sup>Po (half-life 138 days), was determined in Polish mushrooms by Skwarzec and Jakusik (2003). Among 20 tested both edible and inedible species, the highest mean activity concentration of about 40 Bq kg<sup>-1</sup> DM was observed in *Boletus edulis* and *Leccinum scabrum*, while the lowest one, 2.1- 4.3 Bq kg<sup>-1</sup> DM, in *Xerocomus badius* and *X. subtomentosus*, both being typical radiocesium accumulators (see below). This is interesting because all these species are taxonomically closely related. Higher <sup>210</sup>Po levels were reported for caps than for stipes. Despite the high dose coefficient (1.2 x 10<sup>-6</sup> Sv per Bq), even at high *B. edulis* consumption of 5 kg (fresh matter) per year, an effective dose would be 37  $\mu$ Sv. Levels of <sup>210</sup>Po in mushrooms are considerably higher than those in foods of both plant and animal origin.

Activity concentrations of <sup>210</sup>Po higher for one order of magnitude reported Vaaramaa et al. (2009) in Finnish mushrooms. They also observed the highest levels in Boletaceae family, namely in *Leccinum vulpinum*, while the lowest in Russulaceae family. A higher accumulation was proved in caps than in stipes. Yearly radiation doses from <sup>210</sup>Po ingested from mushrooms in Finnish conditions were reckoned up to about 4  $\mu$ Sv.

Rarely reported data on <sup>226</sup>Ra in mushrooms indicate low activity concentrations in France (Kirchner and Daillant, 1998) and even in a Romanian uranium mining area (Popa et al., 2010).

Radionuclide <sup>7</sup>Be was detectable only in a few of 72 samples of various Spanish mushroom species (Baeza et al., 2004).

Overall,<sup>40</sup>K is the main natural radionuclide of edible mushrooms.

Species	Activity	Year(s) of collection	Country	Reference
Xerocomus badius	0.50 - 1.48	1996-1998	Poland	Malinowska et al., 2006
(28 pooled samples)				
12 species	0.59 - 2.02	2002	Turkey	Karadeniz and Yaprak, 2010
11 species	0.72 - 1.78		Turkey	Turhan et al., 2007
10 species	0.8 - 1.8	2004	8 European countries	Szántó et al., 2007
70 species (including	mostly	2006, 2007	Southern Poland (site of a	Mietelski et al., 2010
inedible)	1.0 - 2.5		hot spot of Chernobyl	
			fallout)	

Table 1. Activity concentrations of natural isotope <sup>40</sup>K (kBq kg<sup>-1</sup> DM) in wild growing mushrooms

## Table 2. Activity concentrations of natural isotope <sup>210</sup>Pb (Bq kg<sup>-1</sup> DM) in wild growing mushrooms

Species	Activity	Year(s) of collection	Country	Reference
18 species	<1.8 – 36.5 mean 12.0	1991-1997	France	Kirchner and Daillant, 1998
Xerocomu badius		1996-1998	Poland	Malinowska et al., 2006
(80 pooled samples)				
caps	1.8 - 36.4			
stipes	1.1 - 26.2			
32 species (including	0.8 - 202	1999-2001	Spain	Guillén et al., 2009
inedible)	median 16.6		-	
17 species (including		2006, 2007	Finland	Vaaramaa et al., 2009
inedible)				
caps	1.6 - 16.2			
stipes	1.0 - 15.5			

## 5. ARTIFICIAL RADIONUCLIDES IN MUSHROOMS UNTIL 1985

Several radionuclides were discharged into the global environment through nuclear weapons testing until 1963. The total release of the most important radionuclide, <sup>137</sup>Cs, was estimated as  $9.6 \times 10^{17}$  Bq (for this section, see Kalač, 2001 and references therein).

Most mushroom species have limited abilities to accumulate non-radioactive isotopes of cesium. The reported bioaccumulation factors fruit body/underlying soil of the non-radioactive Cs are not significantly different from those for vascular plants. However, in radiocesium from the fallout, the observed values were higher for at least one order of magnitude.

Limited data were reported for mushroom radioactivity until 1985 in central Europe. Activity concentrations of  $^{137}$ Cs below 1,000 Bq kg<sup>-1</sup> DM were usual. *Xerocomus badius* and *X. chrysenteron* were already then identified as the accumulating species.

Mushrooms did not accumulate <sup>90</sup>Sr or radioisotopes of plutonium at toxicologically significant levels.

## 6. MUSHROOM RADIOACTIVITY AFTER THE CHERNOBYL ACCIDENT

#### 6.1. Chernobyl Accident

The disaster of the Chernobyl (Ukraine) nuclear power station on 26 April 1986 released into the environment about 3.8 x 10<sup>16</sup> Bq from <sup>137</sup>Cs decay. The ratio <sup>137</sup>Cs to <sup>134</sup>Cs was approximately 2 : 1. The main radionuclides produced during an explosive fission reaction are <sup>137</sup>Cs and <sup>90</sup>Sr with long half-lives 30.17 and 28.8 years, respectively. Radiocesium <sup>134</sup>Cs (half-life 2.06 years), also important for mushroom contamination, has been produced in reactors during long-term fission. Moreover, <sup>144</sup>Ce, <sup>131</sup>I, <sup>95</sup>Nb, <sup>239</sup> Pu, <sup>240</sup>Pu, <sup>103</sup>Ru, <sup>106</sup>Ru, <sup>230</sup>Th, <sup>232</sup>Th and <sup>95</sup>Zr were detected in mushrooms early after the accident (Mietelski et al., 2002; Zarubina, 2004). Nevertheless, their toxicological risk from mushroom consumption was limited.

The contamination levels of an area were affected by direction and speed of radioactive cloud, distance from Chernobyl (here and hereafter is meant the power station), and particularly by rainfall or only dry depositions during passage of the clouds. Fallout levels were thus very different even in relatively adjacent sites. For instance, Bakken and Olsen (1990) observed in Norway up to fivefold differences even within squares 20 x 20 cm. Contamination levels varied very widely in the orders of  $10^{1}$ - $10^{5}$  Bq m<sup>-2</sup>. In the heavily polluted areas adjacent to Chernobyl, <sup>137</sup>Cs radioactivity may be of a health concern for up to next three centuries.

During the initial years after the accident, both radiocesium isotopes participated in mushroom radioactivity. Later, since about mid-1990's, <sup>137</sup>Cs has remained the crucial radionuclide.

#### 6.2. Radiocesium Transfer From Soils to Mushrooms

Edible mushrooms have been collected mostly in forests, which present semi-natural ecosystems. Forest soils differ from agricultural cultivated lands. Temperate forest soils are multi-layer with forest floor, hemi-organic and mineral layers. Fungal and microbial activities likely contribute considerably to the long-term retention of radionuclides in organic layers of forest soils. A high variability is thus commonly observed in radionuclide transfers and redistribution patterns in contaminated forests. Soil compartments represent the major reservoir of radionuclides, which can cause long-term contamination of the food chain. Organic matter of forest soils has a lower affinity for cesium than mineral components. Cesium is thus easily available for mushroom mycelium located in organic layers. Long lasting availability of some radionuclides was shown to be the source of much higher transfer in forest ecosystems than in agricultural lands (Calmon et al., 2009).

Contamination levels of <sup>137</sup>Cs observed in years following the Chernobyl accident even within the same species show both high spatial and temporal variability. Several factors have been implicated: mycelium habitat and depth, forest type – fruit body location, sampling strategy, soil clay content and soil moisture and/or microclimate (Gillett and Crout, 2000 and references therein).

Depth of mycelium is a very weighty factor affecting initiation and level of mushroom radioactivity. In species with superficial mycelium (e.g. of genera *Collybia* and *Clitocybe*), the contamination attained within a few months of fallout whilst other deeper penetrating species (e.g. *Boletus edulis*) achieved contamination peaks several years after the deposition. Therefore, ecological half-lives can be estimated with difficulty (Gillett and Crout, 2000).

Transfer of a radionuclide from soil to mushroom fruit body has been usually expressed as "aggregated transfer coefficient" ( $T_{ag}$ ), defined as the ratio between mushroom activity at time *t* and the initial deposit of the radionuclide at time t = 0 (for post-Chernobyl <sup>137</sup>Cs assumed to 1<sup>st</sup> May 1986). The parameter must be decay-corrected to the time of a mushroom sampling. A simplified parameter has been transfer factor (TF; or transfer ratio) defined as the ratio of mushroom activity (Bq kg<sup>-1</sup>) to underlying soil deposit (Bq m<sup>-2</sup>), both at time *t*. Practical differences between TF and T<sub>ag</sub> are relatively small compared to the effects of other factors. Both the parameters are usually expressed as m<sup>2</sup> per kg DM of a mushroom.

Nevertheless, estimation of credible TF values requires consideration of the localization of the mycelia within the soil column. Sampling the whole organic layer can cause overestimation or underestimation of the transfer ratios (Karadeniz and Yaprak, 2007). As reported Baeza et al. (2005) for Spanish pine forests, mushroom mycelium is generally localized in the surface layer of soil (0-5 cm). However, different soil horizons were sampled and analyzed (e.g. 0-2, 0-5, 0-10 cm) by various authors and the reported transfer factors are thus hardly comparable.

Transfer factors for <sup>137</sup>Cs and stable <sup>133</sup>Cs determined in 1998 and 2002 in a scale of mushroom species were fairly constant both in Belorussian (Yoshida et al., 2004) and Turkish forests (Karadeniz and Yaprak, 2007), respectively, with different level of <sup>137</sup>Cs contamination. Thus, some 15 years after the contamination, the studied mushroom species did not distinguish between stable <sup>133</sup>Cs and <sup>137</sup>Cs from Chernobyl. <sup>133</sup>Cs could be thus partially used for the evaluation of <sup>137</sup>Cs behavior.

There exists a consensus of various authors, that mushrooms of different nutritional strategy accumulate <sup>137</sup>Cs in the order mycorrhizal > saprotrophic > or  $\approx$  parasitic (overall in Gillett and Crout, 2000).

Mushrooms showed a greater preference for rubidium and potassium than for cesium. The uptake of <sup>137</sup>Cs could be thus prevented by providing additional K or Rb at contaminated sites (Vinichuk et al., 2010).

Transfer factors of <sup>137</sup>Cs for mushrooms from coniferous forests usually exceeds the factors for mushrooms in deciduous forests (Vinichuk and Johanson, 2003 and references therein).

Until now, most of the <sup>137</sup>Cs in forest soils appears to be available for uptake and radioactive decay likely remains the main factor for a reduction of the radionuclide in forest ecosystems. Several models for <sup>137</sup>Cs migration in forest ecosystems after the Chernobyl accident were developed, however, they have a very high degree of divergence of predictions, particularly when forecasting edible mushrooms contamination.

Only a limited information is available on transfer of other radionuclides to mushroom fruit bodies. The transfer from soil to mushrooms determined in Spanish forests was low for detected both man-made and natural radionuclides. The efficiencies were ranked as follows:  $^{228, 230, 232}$ Th  $\approx ^{40}$ K  $\geq ^{137}$ Cs  $\geq ^{234,238}$ U  $\approx ^{226}$ Ra  $\geq ^{90}$ Sr  $\geq ^{239+240}$ Pu  $\approx ^{241}$ Am. *Hebeloma cylindrosporum*, known by its high transfer factor for  $^{137}$ Cs, was found as an accumulator of other studied radionuclides and can be thus used as a good bioindicator of the radioactivity of an ecosystem (Baeza et al., 2006a; Baeza and Guillén, 2006).

#### 6.3. Mushrooms Radioactivity Since 1986

Skin and flesh had very similar radiocesium concentrations, while the spore-bearing parts (gills or tubes) had for about 50-100% higher levels (Bakken and Olsen, 1990), in a work of Heinrich (1993) even for about 250% higher. Such information was proved by Baeza et al. (2006b) who separated fruit bodies to cap + gills and stipes. Maximum level of  $^{134}$ Cs was found in mature fruit bodies. Also Malinowska et al. (2006) reported higher activity concentrations in caps than stipes of *Xerocomus badius* and similar phenomenon was observed in various species of several families (Bazała et al., 2005).

High accumulation of radiocesium particularly in caps of *X. badius* has been ascribed to natural pigments badion and norbadion A causing chocolate brown or golden yellow coloration of this and related species (Aumann et al., 1989). These pigments have numerous acid-base functional groups, which are able to bind monovalent cations including  $Cs^+$ . Nevertheless, only about 12 % of the total radiocesium was detected in cap skin (epicutis) of *X. badius*, in proportion to its weight (Neukom and Gisler, 1991).

#### 6.3.1. Mushrooms Radioactivity between 1986 and 2000

Data on mushroom radioactivity during the years following the Chernobyl accident were reviewed by Gillett and Crout (2000), Kalač (2001) and particularly by a comprehensive overview of Duff and Ramsey (2008). The latter review collates data until 2000. Numerous references are available therein and are not thus given here.

Overall, activity concentrations varied very widely. In some mycorrhizal species from several hundreds to above 100,000 Bq kg<sup>-1</sup> DM, in saprotrophic and parasitic species between

a few hundreds and a few thousands  $Bq kg^{-1} DM$ . The EU statutory limit of 12,500  $Bq kg^{-1} DM$  was often substantially surpassed. Commonly consumed species with different rates of radiocesium accumulation are given in Table 3.

Table 3. Selected edible mushroom species with different rates of
radiocesium accumulation

High	Medium	Low
Cantharellus lutescens (M)	Agaricus silvaticus (S)	Amanita rubescens (M)
Cantharellus tubaeformis (M)	Boletus edulis (M)	Armillariella mellea $^{st}$
Hydnum repandum (M)	Cantharellus cibarius (M)	Calocybe gambosa (S)
Laccaria amethystina (M)	Leccinum aurantiacum (M)	Laccaria laccata (M)
Rozites caperata (M)	Leccinum scabrum (M)	Lepista nuda (S)
Russula cyanoxantha (M)	Russula xerampelina (M)	Lycoperdon perlatum (S)
Suillus variegatus (M)		Macrolepiota procera (S)
Xerocomus badius (M)		
Xerocomus chrysenteron (M)		

Nutritional strategy: M ... mycorrhizal, S ... saprotrophic, \* mostly parasitic, but with an adaptable strategy

Very wide differences exist within a species. A unique study was carried out in Poland with 278 samples of highly accumulating and widely consumed *Xerocomus badius*, to cover systematically the whole country area, in 1991. Maps of <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>40</sup>K levels in this species were prepared. The most frequent activity concentrations were 2,000-10,000 and 200-600 Bq kg<sup>-1</sup> DM for <sup>137</sup>Cs and <sup>134</sup>Cs, respectively (Mietelski et al., 1994).

An extremely high, probably record value of  $862,100 \text{ Bq kg}^{-1} \text{ DM}$  was determined in inedible *Paxillus involutus* from a site about 70 km west of Chernobyl (Vinichuk and Johanson, 2003).

Data on <sup>134</sup>Cs were reported only until mid-1990's due to the short half-life of the radioisotope. Nevertheless, traces were detected in some species in Poland even in 2006 and 2007 (Mietelski et al., 2010).

Maximum level of mushroom radioactivity was observed in 1987 and mainly in 1988. This was caused by a subsequent supply of the radionuclides to the upper soil horizon through dropped needles and leaching from needles and bark.

#### 6.3.2. Mushrooms Radioactivity Since 2001

The frequency of original papers with information on radioactivity of mushrooms collected after 2000 markedly decreased as compared with the previous period. All following data deal with <sup>137</sup>Cs activity.

Interesting long-term data reported Mascanzoni (2009) in series of activity concentrations determined between 1986 and 2007 in a site of southern Sweden with high Chernobyl fallout contamination of 35,000 Bq m<sup>-2</sup>. In accumulating *Suillus luteus*, values of the activity, decay-corrected to 1<sup>st</sup> May 1986 to avoid a systematic bias of <sup>137</sup>Cs decay, remained stable at level about 120,000 Bq kg<sup>-1</sup> DM, while in a *Cantharellus* spp. the activity increased about three times during the period. Reasons of the difference were not convincingly explained.

Activity concentrations mostly below 100 Bq kg<sup>-1</sup> DM were determined in 33 samples of 15 species gathered from several sites in the Czech Republic and the Slovak Republic with relatively low Chernobyl fallout, during 2000-2004. *S. luteus* and particularly *X. badius* were out of the common run. The maximum measured level in *X. badius* was 6,260 Bq kg<sup>-1</sup> DM (Dvořák et al., 2006). Similarly, caps of *X. badius*, *S. luteus* and *Tricholoma equestre* collected in northern Poland in 2004, had activities 2,700, 1,880 and 1,180 Bq kg<sup>-1</sup> DM (Bazała et al., 2005).

Mean activity concentration of 18 samples belonging to 10 species, collected in eight European countries from Ukraine to Belgium during 2004, was  $834 \pm 1,290$  Bq kg<sup>-1</sup> DM. Also in this work, *X. badius* samples from Poland and Ukraine had the highest determined activities (Szántó et al., 2007).

A specific situation was reported from the most polluted area of Poland, "Opole region hot spot", where the Chernobyl contamination was up to >60,000 Bq m<sup>-2</sup> in 1993. In over 70 species of both edible and inedible mushroom species, collected during 2006 and 2007, a very wide range between 30 and 54,070 Bq kg<sup>-1</sup> DM was determined. Two families showed extraordinary high means, Suillaceae (nearly 12,000 Bq kg<sup>-1</sup> DM) and mycorrhizal species of Tricholomataceae (about 10,000 Bq kg<sup>-1</sup> DM). Commonly, mycorrhizal species had higher activity concentrations than saprotrophic ones (Mietelski et al., 2010).

Quite different situation was reported from Spain with minimum Chernobyl fallout (Baeza et al., 2004). Median of 72 samples belonging to 49 species was only 3.44 Bq kg<sup>-1</sup> DM with accumulating genera *Hebeloma*, *Tricholoma* and *Lactarius*. Similarly as in other reports, mycorrhizal species accumulated more <sup>137</sup>Cs than saprotrophic ones. Unfortunately, period of mushroom sampling was not given.

In conclusion, mushroom contamination with <sup>137</sup>Cs during the decade since 2001 remained relatively stable. The two main recognized factors, level of a site contamination by the Chernobyl fallout and mushroom species, have to be taken under consideration.

## 7. DECREASE OF MUSHROOM RADIOACTIVITY BY CULINARY TREATMENTS

Data on the radioactivity changes during various courses of mushroom preservation have been rare. Only limited information has been available for a decrease of <sup>137</sup>Cs during mushroom soaking and cooking.

The extraction of radiocesium from intact fruit bodies into soaking water or table salt solution is low due to mushroom lipophilic gel-like surface layer. On the contrary, shrinkage between 36-87% of the initial values was reported from boiled slices of several mushroom species. The decrease was higher from dried or frozen slices with disrupted tissue structure than from fresh ones (Steger et al., 1987; Klán et al., 1988; Neukom and Gisler, 1991; Skibniewska and Smoczyński, 1999; Butkus and Dimaviciene, 2009). Naturally, the extracts must be discarded. Such decontamination removes a lot of compounds affecting attractive flavor.

## 8. RADIOACTIVITY BURDEN DUE TO MUSHROOM CONSUMPTION

As results from the previous sections, the calculations of credible radioactivity dose from ingested mushrooms have been rather complicated. Very wide variability of activity levels both among and within species, limited knowledge of radioisotopes bioavailability, likewise fragmentary information on wild mushroom consumption within the population are the main imperfect input data. The reported calculations were carried on for fresh or dried mushrooms and did not take into consideration possible losses of radioisotopes during culinary treatments. Thus, the following values should be appreciated as the qualified estimations.

Using cultivated *Pleurotus eryngii*, Baeza and Guillén (2004) determined very high availability of about 97% of Cs, Sr and Pu, as only about 3% of radioactivity was associated with non-digestible neutral detergent fibre fraction.

As mentioned above, permissible dose limit has been 1,000  $\mu$ Sv year<sup>-1</sup> for adults.

In Bryansk region, the most contaminated area of Russia about 200 km apart of Chernobyl, dose of radiocesium ingested by the rural population from mushrooms was determined to be about 500  $\mu$ Sv per year during 1994-1995 with peaks in fall. Mean annual consumption 5.5 and 9.3 kg of wild mushrooms was used for females and males, respectively (Skuterud et al., 1997). Mushrooms could contribute up to 60-70% of dietary <sup>137</sup>Cs intake of adults during the period 1986-1994 in the region (Shutov et al., 1996).

For mushrooms collected in Poland between 1996 and 1998, the annual incurred effective dose was 80 and 10  $\mu$ Sv for <sup>137</sup>Cs and <sup>210</sup>Pb, respectively. These values were calculated for annual consumption of 10 kg of fresh mushrooms with mean activity concentrations of 5,800 and 13 Bq kg<sup>-1</sup> for <sup>137</sup>Cs and <sup>210</sup>Pb, respectively (Malinowska et al., 2006).

The contribution of 200  $\mu$ Sv to the effective dose was calculated for extreme mushroom consumers in former Czechoslovakia by Klan et al. (1988) for the period immediately after the Chernobyl accident. The input data were 10 kg per year of *X. badius* and *X. chrysenteron* with high mean levels of 10,000 and 2,200 Bq kg<sup>-1</sup> DM of <sup>137</sup>Cs and <sup>134</sup>Cs, respectively. Even though the Chernobyl fallout was relatively low on the territory of Czechoslovakia, mean yearly intake of 1,550 Bq from mushrooms was calculated for 1988. Mushrooms thus became the most significant source of internal contamination (Horyna, 1991).

Markedly lower burden has been in countries with very low Chernobyl fallout together with low consumption of wild mushrooms. This was reported from the United Kingdom for the period 1994-1996 (Barnett et al., 1999), Spain (Baeza et al., 2004) or Turkey (Karadeniz and Yaprak, 2010). The annual effective doses were extremely low, ranging from <1 to 2  $\mu$ Sv. Thus, natural radioisotopes have been the main sources of the burden under such conditions.

## 9. RADIOCESIUM IN MEAT OF GAME EATING MUSHROOMS

Considerable increase of radioactivity in tissues of animals eating mushrooms was reported particularly from Scandinavian and central European countries in years following Chernobyl accident. These regions were stricken by extensive fallout and had a high density of mushrooms. The contamination was observed both in wild animals, such as roe deer (*Capreolus capreolus*), red deer (*Cervus elaphus*), reindeer (*Rangifer tarandus*) and wild boar

(*Sus scrofa*), and in grazing domestic goats and sheep. Commonly, meat of red deer was less contaminated than that of roe deer or wild boar. Radioactivity levels have often and highly surpassed the limit of 600 Bq kg<sup>-1</sup>.

Time trends of meat radioactivity in following years have been different for ruminants and wild boars. In roe deer and red deer, activity concentrations of <sup>137</sup>Cs are rather low, except for the mushroom season in fall, and continually decreasing with time from the Chernobyl fallout. On the contrary, wild boar meat has retained high level of radioactivity, even increasing in some periods (Strebl and Tataruch, 2007). This is due to different habitat and dietary habits. Roe deer lives usually in a relatively small territory and feeds mostly on less contaminated herbal plants, apart from the mushroom season. Wild boar belongs to omnivores accepting a variety of food and is capable to move over long distances (Semizhon et al., 2009).

Time-dependency of <sup>137</sup>Cs activity concentrations in roe deer meat has steadily decreased since 1987. It mostly declined below the hygienic limit during the mid-1990's and fluctuated round about 400 Bq kg<sup>-1</sup> in the early 2000's in the monitored polluted Austrian and German forests. The time course is a combination of two factors – an exponential <sup>137</sup>Cs decay and a peak in August-December due to mushroom feeding. The fall peak decreases with time (Strebl and Tataruch, 2007; Fielitz et al., 2009). In ten Polish regions, mean levels were considerably lower, 6.3 and 8.7 Bq kg<sup>-1</sup> in meat of roe deer and red deer, respectively, in 2007 (Rachubik, 2008).

During the yearly monitoring between 1990 and 2003, mean <sup>137</sup>Cs concentration in wild boar meat fluctuated round about 2,000 Bq kg<sup>-1</sup> in an Austrian region (Strebl and Tataruch, 2007). In a very comprehensive set of 2,433 wild boars shot in SW Germany during 2001-2003, 21-26 and 1-9.3% of tested meat samples exceeded the limit 600 Bq kg<sup>-1</sup> during summer and winter, respectively. An underground mushroom *Elaphomyces granulatus* (common name deer truffles) was found in significantly higher proportions in stomachs with maximum <sup>137</sup>Cs contamination than in stomachs with low level of the radioisotope. Mean <sup>137</sup>Cs activity concentration of 6,030 Bq kg<sup>-1</sup> fresh matter was determined in the mushroom (Hohmann and Huckschlag, 2005). In another region of south Germany, very wide range of 5 – 8,266 Bq kg<sup>-1</sup> of <sup>137</sup>Cs was observed in meat of 656 wild boars shot between 1998 and 2008 (Semizhon et al., 2009). Only low activity concentrations of <sup>137</sup>Cs, in the order of tens Bq per kg, were reported from Poland (Rachubik, 2008) and the Czech Republic (Dvořák et al., 2010).

In monogastric animals, radiocesium is usually excreted via the kidneys. The rate of radiocesium absorption from the intestines into the blood in domestic pigs reached 90-100%. Retention and excretion have an exponential behavior and biological half-time of the absorbed radiocesium is thus short, about 20-40 days (Stara et al. 1971; Voigt et al., 1989).

The activity of <sup>137</sup>Cs in wild boar meat can be reduced to about one quarter of the original level by repeated leaching in table salt solutions (Dvořák et al., 2008).

Thus, the steadily decrease of meat contamination with  $^{137}$ Cs is expected in roe deer and red deer, whereas great variability and in some regions the preservation of recent overrun of the limit of 600 Bq kg<sup>-1</sup> is supposed for a long time, probably a few decades. While the effective dose from game meat is very low in Poland and the Czech Republic except for hunters (and possibly poachers) and their families, the sale of wild boar meat surpassing 600 Bq kg<sup>-1</sup>, based on a system of hunters self-control, was banned in Germany.

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