Role of Fungi in the Determination of the Radiological Status of Terrestrial Ecosystems

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ABSTRACT

In an attempt to assess the importance of the role of fungi as bioindicators in terrestrial ecosystems, we compiled the results reported by more than 100 authors worldwide. These mostly were centred on the 137Cs and 40K contents. The genera Paxillus, Xerocomus, Cantharellus, and Hebeloma were among those with the highest radiocaesium contents. Studies in countries not highly contaminated in the Chernobyl accident, which included other anthropogenic and natural radionuclides in the assays find the order of relative accumulation to be: 40K > 137Cs > 226,230,232Th > 228Ra > 167234,238U > 167226Ra > 16790Sr >> 239+240Pu > 241Am. Laboratory studies showed that the relative position of anthropogenic radionuclides (mainly radiocaesium and radiostrontium) depends on the moment at which fallout occurred. In particular, if the radionuclide deposition occurred long before the formation of the fruiting bodies, the accumulation of radiocaesium was higher than that of radiostrontium. Other factors, such as the concentration of stable elements and the bioavailability of the different radionuclides in the soil, also affect the radioactive content of the fruiting bodies and hence the dose due to their consumption. While the dose from ingestion of mushrooms in areas clearly affected by the Chernobyl Nuclear Power Plant accident is currently estimated at 1.8 mSv/year due to 137Cs, the maximum dose in unaffected areas estimated on the basis of the present findings is 4.60·10^{-3} mSv/year for the main natural and anthropogenic radionuclides present in the environment.

Keywords: anthropogenic radionuclides, dose, mushroom, natural radionuclides, radiocaesium

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INTRODUCTION

As a consequence of the atmospheric nuclear weapon tests carried out from the end of World War II until around the mid-1960s, and of accidents involving nuclear materials such as Thule, Palomares, Chernobyl, and the re-entry of SNAP-9 (UNSCERAR, 1982), large quantities of anthropogenic radionuclides have been released into the environment. They can be transferred along the trophic chain and finally incorporated by humans, with the resulting implied health risk. Therefore, their content and transfer in different products – basically milk, meat, fish, vegetables, and their derivatives – have frequently been analyzed (Ng 1982). However, little attention was paid in these studies to the consumption of mushrooms. We consider that there may have been two reasons. On the one hand, mushrooms are strongly seasonal, appearing mainly in autumn and to a lesser degree in spring. This, together with their short life span led to the inference that they did not constitute a part of fresh market produce during a significant part of the year. And on the other hand, since mushrooms are consumed mainly by only a small number of people, usually of rural origin, it was supposed that the dose due to mushroom ingestion might be less than that of other produce.

However, after the Chernobyl Nuclear Power Plant accident in 1986, there was an increase in the number of studies of the radioactive contamination in mushrooms. Their focus was mainly on 137Cs, and to a lesser extent on 90Sr. These radionuclides are usually considered to be indicators of radioactive contamination risk. Both are products of nuclear...
fission, with significant production rates and relatively long half-lives, and they are chemical analogues of biologically important cations – potassium and calcium, respectively. From these studies, two surprising results arose. The radionuclide content, especially $^{137}$Cs, of mushrooms was higher than that of other food items collected in the same area (Horyna 1991; Ruig et al. 1992; Skuterud et al. 1997), and generally higher than other forest products such as bilberries (Mietelski et al. 1996; IAEA 2006). In a review of the radioactive contamination in the Arctic (Alaska, Artic Finland, Artic Norway, Artic Sweden, Greenland, Iceland, Northern Canada, Northern Russia), Strand et al. (2002) found that the order of the level of $^{137}$Cs contamination in products harvested in natural and semi-natural ecosystems was: Reindeer (~800 Bq/kg), mushrooms (~300 Bq/kg), freshwater fish (~50 Bq/kg) > lamb meat, goat cheese (~100-200 Bq/kg) > potatoes, vegetables (~3-9 Bq/kg) > marine products (~0.1 Bq/kg).

Although the dietary ingestion of mushrooms was estimated at 7.8%, they presented the second highest $^{137}$Cs content, only surpassed by reindeer meat.

In addition, the ecological half-life of radionuclides in mushrooms is longer than in other products (Shutov et al. 1996; Skuterud et al. 1997). This may explain the annual increase in some compartments of the trophic chain in highly contaminated areas (Shutov et al. 1996; Skuterud et al. 1997). Their consumption may even be a significant percentage of people’s annual dose (Mietelski et al. 1994; Shutov et al. 1996; Skuterud et al. 1997).

Another pathway of incorporation to man is through the consumption of mushrooms by animals. Several studies have found a relationship between the consumption of mushrooms by ruminants, and the presence of radionuclides in their meat or milk. There have also been observed seasonal variations in the body content of radio caesium in these animals due to the seasonal availability of mushrooms and lichens (Hove et al. 1990; Strandberg et al. 1994a; Mehl et al. 1998; Ávila et al. 1999; Zibold et al. 2001; IAEA 2006).

**ACTIVITY LEVELS IN MUSHROOMS**

**Radio caesium**

In the literature, the radionuclides most extensively studied are two radio caesium isotopes – $^{134}$Cs and $^{137}$Cs. As can be seen in Fig. 1, more than 90% of the studies that we consulted included radio caesium assays. These studies were carried out mainly in northern, central, and eastern European countries, i.e., those that were most affected by the Chernobyl accident. We found only a small number of studies for the Americas (only Canada and Mexico) and Asia (only Japan and Taiwan). No references were found for Africa or Oceania. Table 1 lists the ranges of $^{137}$Cs content in mushrooms collected in different countries around the world, with the species that presented high contents and the corresponding references. The $^{137}$Cs content is usually expressed on the basis of dry weight of mushroom, and only to a lesser extent on the fresh weight. In the latter case, we normalized the activity levels by assuming that the dry weight was about 10% of fresh weight. The most significant fact was the high variability of the accumulation of radio caesium in mushrooms. The $^{137}$Cs content in mushrooms ranged over about eight orders of magnitude, from 0.4 Bq/kg d.w. to 50 MBq/kg d.w. in some areas highly contaminated by the Chernobyl accident. Some studies have found the radio caesium content to vary over about two or three orders of magnitude within mushrooms collected in the same area, and within mushrooms of the same species collected in different locations (Gentili et al. 1991; Mietelski et al. 1994).

In an analysis of the influence of the intraspecific variation of $^{137}$Cs activity concentration in spores of *Suillus variegatus* in seven Swedish locations, Dahlberg et al. (1997) found that 40% of the variance of the $^{137}$Cs content was explained by site variations, but that the other 60% could not be explained by genetic differences.

The distribution of radio caesium content in mushrooms of a given population has been found to follow a non-symmetric, log-normal type of distribution (Mietelski et al. 1994; Zibold et al. 2001; Baeza et al. 2004a). It is therefore advisable not to use mean values and standard deviations to evaluate the degree of radio caesium contamination of mushrooms in an ecosystem. In our opinion, it is better to describe it using the median value and the range observed.

dependence on the nutritional mechanism of the mushroom has also been reported. Mycorrhizae, forming symbiosis with tree roots, present higher $^{137}$Cs contents than saproph ytes or parasites (Guillite et al. 1994; Kammerer et al. 1994; Yoshida et al. 1994b; Baeza et al. 2004a; IAEA 2006).

This factor was considered to be more important than the environmental conditions or the properties of the soil in which the mushrooms were growing (Heinrich 1992). Thus the relative order of radio caesium contamination can be summarized as: Mycorrhiza > saprophyte = parasite.

Some of the species that were found to have high radio caesium contents are listed in Table 1. Most are mycorrhizae such as *Hydnium repandum, Paxillus involutus, Rozites caperata, Xerocomus badius*, and some species of the genera *Hebeloma, Cortinarius, and Lactarius*. Some authors suggest that mycorrhizae act as a filter through which their host plants take up nutrients. In this process, the host plant might discriminate against caesium in favour of potassium (Guillite et al. 1994; Kammerer et al. 1994). This hypothesis was confirmed to some extent in laboratory experiments, with the observation of a decrease in the uptake of $^{134}$Cs by germinated seeds of *Picea abies* colonized by *Hebeloma crustuliniforme* mycorrhiza (Riesen et al. 1996) and an accumulation of $^{134}$Cs in fungal hyphae (Brunner et al. 1996).
although no fruiting body was obtained in these experiments. One species analyzed in most of the studies was *Xerocomus badius*. According to Kotike et al. (1998), it contains a large amount of macro- and micro-elements (P, K, Ca, Mg, Fe, Zn, Mn, and Al) which they take to be connected to activity in the hyphal sheet and to the frequent occurrence of vascular bodies where deposition can occur. It was also found that some pigments present in the cap—derivatives of pulvinic acid, badione A1 and norbadione A2—bind potassium and caesium (Aumann et al. 1994; Smith et al. 1995; Ingrao et al. 1998). But their bioavailability in soils was considered to be different—137Cs pre-Chernobyl was considered to be more fixed to soil particles than “fresh” 134Cs, due to the time that had passed since the global fallout (Muramatsu et al. 1994a, 1994b; Ban-Nai et al. 1993; Tsukada et al. 1994, 1995; Mietelski et al. 1994; Tsvetnona et al. 2003, 2004; Vinichuk et al. 2002; Travnikova et al. 2002; Travnikova et al. 2003, 2004). Therefore the comparison of the ratio 134Cs/137Cs in mushrooms, expressed in Bq/kg d.w., in different countries worldwide, together with the species that showed major accumulations of radioaesium and the corresponding literature references.

**Table 1** Range of $^{137}$Cs content in mushrooms, expressed in Bq/kg d.w., in different countries worldwide, together with the species that showed major accumulations of radioaesium and the corresponding literature references.

<table>
<thead>
<tr>
<th>Country</th>
<th>Species</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>Xerocomus badius, Hydnum repandum, Rozites caperata</td>
<td>Teherani 1987; Teherani 1988; Ismail et al. 1995</td>
</tr>
<tr>
<td>Belgium</td>
<td>Cortinarius brunneus, Cortinarius armillatus, Laccaria amethystina, Paxillus involutus</td>
<td>Andolina et al. 1990; Fraiture et al. 1991; Guillote et al. 1991; Lambinon et al. 1998</td>
</tr>
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<td>Bulgaria</td>
<td>Craterellus cornucopioides</td>
<td>Calmet et al. 1998</td>
</tr>
<tr>
<td>Czech Republic</td>
<td>Laccaria amethystina, Cortinarius armillatus, Xerocomus badius</td>
<td>Randa et al. 1990; Cibulka et al. 1996; Calmet et al. 1998; Dvořák et al. 2006</td>
</tr>
<tr>
<td>Denmark</td>
<td>Rozites caperata, Cortinarius albiviolaceus</td>
<td>Strandberg et al. 1994a; Strandberg 1994b, 2004</td>
</tr>
<tr>
<td>Finland</td>
<td>Hydnum sp., Rozites caperata, Xerocomus badius, Hydnum repandum</td>
<td>Ilkahimonen et al. 2003</td>
</tr>
<tr>
<td>France</td>
<td>Cantharellus sp., Rozites caperata, Xerocomus badius</td>
<td>Calmet et al. 1998; Kiechler et al. 1998</td>
</tr>
<tr>
<td>Germany</td>
<td>Hebeloma sp., Hydnum repandum, Paxillus involutus, Rozites caperata</td>
<td>Rückert et al. 1990; Römelt et al. 1990; Heinrich 1993; Kammeret et al. 1994; Zibold et al. 2001</td>
</tr>
<tr>
<td>Great Britain</td>
<td>Hydnum repandum, Boletus badius, Cantarellus cibarius</td>
<td>Barnett et al. 1999, 2001; Toal et al. 2002</td>
</tr>
<tr>
<td>Hungary</td>
<td>Tricholoma terreum, Suillus granulatus</td>
<td>Vaszari et al. 1992</td>
</tr>
<tr>
<td>Italy</td>
<td>Cantharellus lutescens, Clytoche infundibuliformis</td>
<td>Battiston et al. 1989; Giovannini et al. 1990; Borio et al. 1991; Gentili et al. 1991; Ingrao et al. 1992</td>
</tr>
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<td>Lithuania</td>
<td>Tricholoma equestre, Cantharellus cibarius</td>
<td>Calmet et al. 1998</td>
</tr>
<tr>
<td>Norway</td>
<td>Lactarius sp., Amanita falva, Amanita vaginata, Cortinarius armillatus, Rozites caperata, Xerocomus badius</td>
<td>Randa et al. 1990; Cibulka et al. 1996; Calmet et al. 1998; Dvořák et al. 2006</td>
</tr>
<tr>
<td>Poland</td>
<td>Xerocomus badius, Sarcodon imbricatum</td>
<td>Bem et al. 1990; Mietelski et al. 1993; Grabowski et al. 1994; Mietelski et al. 1994; Zagrodski et al. 1994; Mietelski et al. 1996; Pietrzak-Fils et al. 1996; Calmet et al. 1998; Mietelski et al. 2002; Malinowska et al. 2006</td>
</tr>
<tr>
<td>Romania</td>
<td>Hydnum repandum</td>
<td>Calmet et al. 1998</td>
</tr>
<tr>
<td>Slovakia</td>
<td>Suillus luteus, Russula aeruginae</td>
<td>Čipáková 2004; Dvořák et al. 2006</td>
</tr>
<tr>
<td>Spain</td>
<td>Hebeloma cylindrosporum, Lactarius deliciousius</td>
<td>Arrondo 1988; Paniagua 1991; Baesa et al. 2004a</td>
</tr>
<tr>
<td>Switzerland</td>
<td>Boletus edulis, Rozites caperata, Xerocomus badius</td>
<td>Froidevaux et al. 2001</td>
</tr>
<tr>
<td>Sweden</td>
<td>Dermocybe cinnamomea, Hygrocybe sp., Rozites caperata, Russula decolorans, Suillus variegates, Cortinarius collinitus</td>
<td>Mascalzoni et al. 1994; Smith et al. 1994; Smith et al. 1995; Smith et al. 1997; Nikolova et al. 1997; McGee et al. 2000; Andersson et al. 2001</td>
</tr>
<tr>
<td>Ukraine/Russia</td>
<td>Paxillus involutus, Xerocomus badius, Lactarius torminosus</td>
<td>Hoshi et al. 1994; Smith et al. 1994; Tsvetnona et al. 1994; Lux et al. 1995; Mietelski et al. 2002; Travnikova et al. 2002; Vinichuk et al. 2003, 2004</td>
</tr>
<tr>
<td>Yugoslavia</td>
<td>Cortinarius armillatus, Laccaria amethystana</td>
<td>Calmet et al. 1998</td>
</tr>
<tr>
<td>AMERICA</td>
<td>Leucopaxillus giganteus, Laccaria laccata</td>
<td>Mihok et al. 1989; Smith et al. 1994</td>
</tr>
<tr>
<td>Canada</td>
<td>Leucopaxillus giganteus, Laccaria laccata</td>
<td>Mihok et al. 1989; Smith et al. 1994</td>
</tr>
<tr>
<td>Mexico</td>
<td>Clavariadelphus truncatus, Gomphus floccosus, Cortinarius caesareikens</td>
<td>Goso et al. 1998; Goso et al. 2000</td>
</tr>
<tr>
<td>Taiwan</td>
<td>Ganoderma tsuga, Lentinula edodes</td>
<td>Wang et al. 1998</td>
</tr>
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</table>
lity. The variation was about three orders of magnitude considering studies worldwide (Masczynski et al. 1990; Römmlert et al. 1990; Paniagua 1991; Kozhevnikova et al. 1993; Mietelski et al. 1993; Baetza et al. 2004b, 2006b). The highest 137Sr content found was 311 Bq/kg d.w. for *Paxillus involutus* in the 30 km zone around Chernobyl (Lux et al. 1995). For Poland, Germany, Sweden, Finland, and Spain, the highest activity level was about 4-5.6 Bq/kg d.w. for *Strophariopsis* and *Amanita* species.

The highest activity level was 1872 Bq/kg d.w., detected in *Lycoperdon perlatum* collected in Italy. The ratio 241Am/239+240Pu was less than unity, within the range 0.001-0.70 (Masczynski et al. 1990; Baetza et al. 2004b), indicating that mushrooms are able to accumulate 137Cs better than 137Sr. The strontium content of mushrooms was less than that found in other plants in the same ecosystems (Römmlert et al. 1990; Yoshida et al. 1998). For 106Ru, 144Ce, and 155Eu contents of mushrooms were below detection limit, from 10 2-103 mBq/kg d.w. (Mietelski et al. 1992; Romeo et al. 1994). We found only one reference – Alkhimova et al. 1994). The isotope 110mAg has only occasionally been detected in mushrooms, such as after the Chernobyl accident.

The content of 238,239+240Pu was lower than that of other anthropogenic radionuclides considered. The values of the Pu content ranged over about five orders of magnitude, from 1.2 mBq/kg d.w. to 82.8 Bq/kg d.w. (Mietelski et al. 1993; Mietelski et al. 1994; Lux et al. 1995; Yamamoto et al. 1995; Mietelski et al. 2002; Outola et al. 2003; Baetza et al. 2004b, 2006b). But the range was 0.39-82.8 Bq/kg d.w. considering only the Ukrainian mushrooms. *Cantharellus cibarius* presented the highest Pu activity, followed by *Xerocomus badius*, *Paxillus involutus*, and *Sulphur luteus* (Lux et al. 1995; Mietelski et al. 2002).

In other countries (Finland, Poland, and Spain), the range was lower (1.2-161 mBq/kg), and the species that had high contents were *Heloboloma cylindrosporum*, *Amanita muscaria*, and *Cantharellus badius*. The ratio 239Pu/238Pu reflected the origin of the contamination – for Ukraine samples it was about 0.5, confirming their Chernobyl origin, and lower for samples from other countries less affected by the Chernobyl fallout. The 214Am content of mushrooms has only been analyzed in Finland and Spain (Outola et al. 2003; Baetza et al. 2006b). The range was 5.4-40 mBq/kg d.w., with *Clitocybe sp.*, *Heloboloma cylindrosporum*, and *Lycoperdon perlatum* presenting the highest accumulations. In general, the ratio 214Am/228Th was less than unity.

In a few studies carried out after the Chernobyl accident, other anthropogenic radionuclides were detected in mushrooms (Battiston et al. 1989; Romeo et al. 1994; Lux et al. 1995). 108Ru, 140Ce, and 152Eu contents of mushrooms were below the detection limit. 108Ru was detected in Austria, Italy, and Ukraine in the range 10-1480 Bq/kg d.w., with *Macropleiota procerca*, *Cantharellus sp.*, and *Clitocybe infundibuliformis* presenting the highest activity levels (Teherani 1987; Battiston et al. 1989; Romeo et al. 1994). For 110mAg, we found only one reference – *Amanita rubescens* with 1.1 - 44.4 Bq/kg (Romeo et al. 1994).

### Natural origin radionuclides

Of the radionuclides of natural origin, 40K is the most extensively studied in the literature (see Fig. 1) (Muramatsu et al. 1991; Vaszari et al. 1992; Mietelski et al. 1993; Wang et al. 1998; Baetza et al. 2004a). The range of variation of 40K content in mushrooms was not wide: 10-100 Bq/kg d.w., independently of the species considered or the location in which the mushrooms were collected. The frequency distribution for 40K is symmetric and almost Gaussian, differing significantly from that of radioactinium (Mietelski et al. 1994; Baetza et al. 2004a). It was also observed that 40K content did not depend on the nutritional mechanism of the mushroom – mycorrhizae and saprophytes had the same mean values and standard deviations (Yoshida et al. 1994; Baetza et al. 2004a).

The highest activity level was 5880³.3 – 11.3 Bq/kg d.w. (Yamamoto et al. 1995; Mietelski et al. 2002; Baetza et al. 2004b; Jia et al. 2004). The highest activity was found in Kosovo, former Yugoslavia (Jia et al. 2004), but unfortunately the species was not mentioned. The maximum content of uranium in mushrooms collected in Spain was 7.0 Bq/kg d.w. for *Amanita ponderosa*, followed by *Russula cossans* and *Heloboloma cylindrosporum* (Mietelski et al. 2002; Baetza et al. 2004b). In Poland, the maximum activity was 240 mBq/kg d.w. in *Macropleiota procerca*, followed by *Xerocomus badius* (Mietelski et al. 2002). The 235U/238U ratio was around unity so that the two isotopes were in equilibrium in these mushrooms, and the 235U/238U ratio confirmed the natural origin of the uranium. In Kosovo, 234U was also detected in one mushroom sample, with the 235U/238U ratio confirming the presence of depleted uranium in that species (Jia et al. 2004).

Thorium isotopes in mushrooms have only been analyzed in Poland and Spain (Mietelski et al. 2002; Baetza et al. 2004b). The 228Th, 229Th, and 230Th contents varied within the ranges 1.8-13 Bq/kg d.w., 2.5·10⁻²-6.5 Bq/kg d.w., and 2.5·10⁻¹-10.7 Bq/kg d.w., respectively. The activities were in the order 228Th > 229Th > 230Th. The higher levels of 228Th relative to 232Th were probably due to the uptake of 228Ra which later decayed to 228Ac and then to 228Th (Baetza et al. 2004b). A clear difference was observed between mushrooms from Poland and Spain. The 230Th and 232Th contents in Polish mushrooms were of the order of mBq/kg d.w., and 228Th was not measured. Spanish mushrooms presented higher activity levels, up to the order of Bq/kg d.w., with the highest contents being in *Amanita ponderosa*, *Russula cossans*, *Tricholom sp.*, and *Heloboloma cylindrosporum* for 228Th and 232Th, and *Amanita ponderosa*, *Russula cossans*, and *Macropleiota procerca* for 230Th.

The radium content in mushrooms was 0.089-87 Bq/kg d.w. (Paniagua 1991; Yamamoto et al. 1995; Kirchner et al. 1998; Baetza et al. 2004a). *Terfezia arenaria*, *Lycoperdon perlatum*, and *Russula cossans* presented the highest radium activity levels. The radium content distribution was probably symmetrical and of a Gaussian type. There was no observed dependence of the radium content on the nutritional mechanism of the mushrooms (Baetza et al. 2004a).

The 210Pb content in mushrooms was in the range 3.64-58.9 Bq/kg d.w. (Kirchner et al. 1998; Malinowska et al. 2006). *Boletus edulis*, *Xerocomus badius*, and *Clitocybe nebularis* presented the highest activities. Kirchner et al. et al. (1998) found that the 210Pb content in mushrooms was greater than that of 226Ra in the same sample. The 210Pb mainly originated from direct uptake from the soil. Deposition of atmospheric 210Pb onto the fruiting bodies and uptake of 222Rn in solution in the soil pore water which subsequently decayed into 210Po were the main radioactivity sources. The radon activity distribution was probably symmetrical and of a Gaussian type. There was no observed dependence of the radium content on the nutritional mechanism of the mushrooms (Baetza et al. 2004a).
The $^7$Be content was in the range 1.5–70 Bq/kg d.w., and the frequency distribution was of log-normal type. There was observed a dependence on the nutritional mechanism, with the content of saprophytes being higher than that of mycorrhizae.

**Comparison of natural and anthropogenic radionuclides**

In the previous sections we have analyzed the range of variation of natural and anthropogenic radionuclides worldwide. But it is also very useful to compare the activity levels of these radionuclides within an ecosystem. 

*Table 2* presents a summary of the ranges of the activities detected. Since the content of natural radionuclides other than $^{40}$K has not been extensively studied, we only considered Spanish and Polish samples in which the uranium and thorium contents were also analyzed. The order of accumulation of radionuclides, considering mean and median values, was the following:

a. Spanish mushrooms, as an example of a region mainly affected by global fallout: $^{40}$K > $^{137}$Cs > $^{228,230,232}$Th ≈ $^{224,226}$Ra > $^{239+240}$Pu > $^{241}$Am

b. Polish mushrooms, as an example of a region highly contaminated by the Chernobyl accident: $^{137}$Cs > $^{40}$K > $^{90}$Sr >> $^{230,232}$Th/g167 $^{234,238}$U/g167 $^{239+240}$Pu/g167

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Range (Bq/kg d.w.)</th>
<th>Natural origin</th>
<th>Radionuclide</th>
<th>Range (Bq/kg d.w.)</th>
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<tr>
<td>$^{137}$Cs</td>
<td>10$^{-1}$ – 10$^{2}$</td>
<td>$^{137}$Cs</td>
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<td>$^{90}$Sr</td>
<td>10$^{-2}$ – 10$^{2}$</td>
<td>$^{222}$Rn</td>
<td>$^{131}$I</td>
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<td>$^{110m}$Ag</td>
<td>10$^{-1}$ – 10$^{2}$</td>
<td>$^{226}$Ra</td>
<td>$^{134}$Cs</td>
<td>10$^{-1}$ – 10$^{7}$</td>
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<tr>
<td>$^{238}$, $^{239+240}$Pu</td>
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<td>$^{236}$U</td>
<td>$^{237}$Np</td>
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<td>$^{237}$Np</td>
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<td>$^{232}$Th</td>
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<td>$^{231}$Pa</td>
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<td>$^{210}$Po</td>
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<td>$^{125}$Sb</td>
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<tr>
<td>$^{103}$Ru</td>
<td>10$^{-1}$ – 10$^{1}$</td>
<td>$^{232}$Th</td>
<td>$^{232}$Th</td>
<td>10$^{-3}$ – 10$^{1}$</td>
</tr>
<tr>
<td>$^{131}$I</td>
<td>10$^{-1}$ – 10$^{1}$</td>
<td>$^{210}$Po</td>
<td>$^{210}$Po</td>
<td>10$^{-1}$ – 10$^{1}$</td>
</tr>
</tbody>
</table>

**FACTORS AFFECTING THE RADIONUCLIDE CONTENT**

The radionuclide content in mushrooms can be influenced by several factors including (i) the date at which radioactive fallout occurred, (ii) its quantity and chemical form, (iii) the location of the layer of soil from which the mycelium takes up nutrients, (iv) the presence of stable elements, and (v) the stage of development of the mushrooms.

**Influence of the source term**

The predominant source term of radionuclides in a given ecosystem was the most significant factor influencing their content in mushrooms. For the anthropogenic radionuclides, especially radioaescaesium, a clear difference is observed between the content in countries strongly affected by Chernobyl fallout (Ukraine, Russia, Sweden, Finland, Poland, etc.) and others in which the predominant source was global fallout (Canada, Spain, Mexico). In the former, the order of magnitude of the radionuclide content was up to 10$^2$ Bq/kg d.w., and in the latter it was up to 10$^2$ Bq/kg d.w. This factor also affected the kind of anthropogenic radionuclides detected. In the case of Chernobyl fallout, short half-life radionuclides such as $^{110m}$Ag, $^{125}$Sb, $^{103}$Ru, $^{131}$I, were detected, while for the case of global fallout only radionuclides with longer half-lives, such as $^{137}$Cs, $^{90}$Sr, $^{239+240}$Pu, and $^{241}$Am, were detectable.

The time elapsed between the incorporation of anthropogenic radionuclides to the ecosystem and the appearance of the fruiting bodies also influenced the radionuclide content. This was analyzed in laboratory experiments under controlled conditions, and dependence of the transfer of $^{134}$Cs and $^{85}$Sr on the time lapse between the incorporation of the radionuclides and harvesting the fruiting bodies. Harvest was taken as time zero. In Exp. B, $^{134}$Cs and $^{85}$Sr were incorporated into the substrate 20 days before the inoculation with mycelium. In Exp. C, they were incorporated at the time of mycelium inoculation, 55 days before harvest. In Exps. D1, D2, and D3, they were incorporated on the surface soil at the different stages of growth of the fungus shown. Modified from Baeza et al. (2002).

**Fig. 2 Time-line of the experiments on Pleurotus eryngii culture grown under controlled laboratory conditions,** and dependence of the transfer of $^{134}$Cs and $^{85}$Sr on the time lapse between the incorporation of the radionuclides and harvesting the fruiting bodies. Harvest was taken as time zero. In Exp. B, $^{134}$Cs and $^{85}$Sr were incorporated into the substrate 20 days before the inoculation with mycelium. In Exp. C, they were incorporated at the time of mycelium inoculation, 55 days before harvest. In Exps. D1, D2, and D3, they were incorporated on the surface soil at the different stages of growth of the fungus shown. Modified from Baeza et al. (2002).

**Stage of maturity of fruiting bodies**

There are few references in the literature discussing the influence of the stage of maturity on the radionuclide content. This was analyzed in laboratory experiments under controlled conditions for the uptake of $^{134}$Cs and $^{85}$Sr by fruiting bodies of *Pleurotus eryngii* (Baeza et al., 2000, 2002). Fig. 2 shows the time-line of the inoculation with the $^{134}$Cs and $^{85}$Sr contaminants at different stages of growth of the fungus. The results of the transfer to the fruiting bodies were evaluated by the transfer coefficients, TC in the figure, and the coefficients λ as calculated from a compartmental model. Both TC and λ were represented as a function of the elapsed time between contamination and harvest. It was found that the longer this elapsed time, the less $^{134}$Cs and $^{85}$Sr the fruiting bodies accumulated. It was also observed that if the radionuclides were incorporated a few days before harvest the accumulation of $^{85}$Sr was greater than $^{134}$Cs, probably due to a change in nutritional requirements. This was not observed in field conditions from global fallout or the Chernobyl accident, because the latter occurred in April 1986, several months before autumn. In these studies, together with this elapsed time, different uptake routes were considered – contamination of the mycelium, contamination of the surface soil, and direct deposition onto the fruiting bodies. The most effective incorporation route was the direct deposition onto the fruiting bodies, followed by soil and mycelium contamination for $^{134}$Cs and these two in reverse order for $^{85}$Sr.
young and old mushrooms presented the same activity levels for $^{85}$Sr, but the old mushrooms presented a greater accumulation of $^{134}$Cs than the young ones.

**Location of the mycelium**

The location of the mycelium is a factor that significantly affects the radionuclide content in mushrooms. The radionuclide content of the soil layer from which the mycelium takes up nutrients seemed to be one of the most important factors controlling the content in the fruiting bodies. Wang et al. (1998) and Ban-Nai et al. (1997) observed that artificially cultivated mushrooms had a lower radiocaesium content than wild mushrooms due to the lower content in the medium used for growth — usually sawdust or rice bran. Yoshida et al. (1994a) also found differences between mushrooms with the mycelium growing in different habitats — wood, litter layer, surface soil layer (0-5 cm), and deeper layers of soil (>5 cm).

There are different methods of locating the mycelium in the soil. The first is the mechanical separation of the soil into different fractions such as rhizosphere, the root-soil interface, the mycelium – has the consequence that the type of ecosystem received a recent deposition of $^{134,137}$Cs significantly greater than that of the 137Cs already present in the soil, such as occurred in certain regions following the Chernobyl NPP accident. This method could be questioned because it is based on the assumption that $^{137}$Cs and $^{137}$Cs have the same bioavailability in the soil (Muramatsu et al. 1991). And a third method is to estimate the fungal biomass by evaluating some organic compound present in the mycelium. Frequently ergosterol is used. It is the main sterol present in most fungi being a principal component of their cell walls, while it is absent from practically all other life forms, in particular from the roots of plants (Weete 1974). The disadvantage of this method is that it does not allow the species of fungi to be identified, so that it can only give an average of the distribution of mycelium at a certain depth of soil.

Olsen et al. (1990) estimated the fungal biomass in a mountain area of Norway to be in the range 12-138 mg fungal d.w./g soil d.w., which was able to retain about 32% of the radiocaesium present in the soil. Nikolova et al. (2000) found the highest concentrations of $^{137}$Cs, as well as the highest organic content, to be in the soil-root interface fraction of small roots and mycelium. Vinichuk et al. (2003) estimated the fungal biomass in soil to be about 13.8 mg/g soil, and to retain 0.1-50% (mean 15%) of the total soil activity of $^{137}$Cs. They also found a higher concentration of $^{137}$Cs in the fruiting bodies than in the mycelium, with ratios in the range 0.1-66 (mean 9.9).

**Bioavailability of radionuclides in soil**

The bioavailability of a given radionuclide is defined as the fraction which is capable of being transferred. The main problem which arises with this concept is that there is no unique operative definition of the bioavailable fraction. In practice, each researcher uses an ad hoc definition that may vary according to the extraction scheme. The most common extractants are ammonium acetate and MgCl$_2$. Fig. 3 shows, by way of example, the speciation of several natural and anthropogenic radionuclides (mainly due to global fallout) radionuclides in a pinewood forest ecosystem. From this figure it is possible to deduce that, with respect to the anthropogenic radionuclides, $^{85}$Sr was commonly observed to be the most mobile (Knatko et al. 1996; Krouglov et al. 1998; Baeza et al. 2005). The mobility of $^{137}$Cs was lower and depended on the presence of clay in the soil (Ohnuki 1994). Humic substances in the soil, however, could reduce clay's absorption of radiocaesium (Dumat et al. 1999). $^{238}$Pu and $^{239}$Pu and $^{239}$Am were more mobile than radiocaesium in a pinewood ecosystem (Baeza et al. 2006b). This was probably due to the strong association of these radionuclides to the organic matter in the soil (Fujikawa et al. 1999). Natural radionuclides were generally strongly bound to soil particles, because they formed an integral part of them (Baeza et al. 2005b, 2006a). $^{226}$Ra was the most mobile natural radionuclide, followed by the uranium isotopes. $^{9}$K and thorium isotopes were the least mobile radionuclides — more than 90% of their activities were strongly attached to soil particles or associated with the residual fraction.

Some workers (Heinrih 1992; Gerzabek et al. 1998) have reported that a combination of these last two properties — bioavailability of the radionuclides and location of the mycelium — has the consequence that the type of ecosystem in which the fungi are growing affects their radiocaesium activity levels. The order of the incorporation is: Coniferous forest > deciduous leaf forest > prairies.

**Relationship with stable elements**

The content of different stable cations in mushrooms did not vary over a wide range, indicating that some of them were essential to the fungi’s development (Ismail et al. 1995). Occasionally, a positive correlation was found between one stable cation and $^{137}$Cs (Ismail et al. 1995), but this depended on the species of mushrooms considered, and was not general.

The content of alkali and alkaline earth elements in mushrooms was different from that found in plants of the same ecosystem (Yoshida et al. 1998). The values were in the following order:

a) Mushrooms: K > Mg > Na > Ca > Rb > Ba > Cs > Sr
b) Plants: Ca > K > Mg > Na > Sr > Ba > Rb > Cs

Compared to plants, the elemental composition of mushrooms was characterized by high Rb and Cs contents and low Ca and Sr contents. Various studies found $^{137}$Cs and stable Cs to be correlated (Randa et al. 1990; Ingrao et al. 1992; Yoshida et al. 1998; Tsukada et al. 1998). Yoshida et al. (1998) found that the ratio $^{137}$Cs/Cs in mushrooms was higher than that in the soil. Subsequent research (Dragović et al. 2003; Yoshida et al. 2004) found that the $^{137}$Cs/Cs ratio was fairly constant for each sampling site, and of the...
same order of magnitude as that in the organic layer of soil, suggesting that \(^{137}\)Cs from Chernobyl was well mixed with stable caesium within the biologically connected compartments in the forest ecosystem. There was no correlation between the K and Cs content in mushrooms: the K content seemed to be species independent, while the Cs content varied widely. This suggested that Cs and K play different roles in mushrooms. The behaviour of Rb was intermediate: Rb was correlated with Cs, but the Rb/Cs ratio was not constant, but decreased with increasing Cs content. This suggested that Rb was partially incorporated by the same mechanism as Cs.

Producing the fruiting bodies of mushrooms under controlled laboratory conditions is very difficult. It has only been achieved with a few species, mainly saprophytes such as \(\textit{Pleurotus eryngii}\). Instead, some studies have analyzed the uptake of radionuclides and the influence of different stable elements using hydroponic cultures of the mycelium of various fungi species. Studies with the mycelium of the \(\textit{Pleurotus}\) genus found a decrease in the stable Cs and \(^{137}\)Cs content when the Cs content was increased in the culture medium (Kuwahra et al. 1998; Terada et al. 1998; Baeza et al. 2005).

**DOSE DUE TO MUSHROOM CONSUMPTION**

The consumption of mushrooms was found to make a significant contribution to radiocaesium incorporation in human body (Shutov et al. 1996; Skuterud et al. 1997). In some countries such as the Czech Republic it was assessed to be the greatest source of radiocaesium uptake in the diet (Ho-ryna 1991). The dose from internal exposure was 0.2-5.35 mSv/y (Shutov et al. 1996; Beresford et al. 2001; Jacob et al. 2001; Dragovic et al. 2003).

One of the main problems connected with the consumption of mushrooms is that, in zones affected by Chernobyl, the dose due to the consumption of wild and farm produce has fallen drastically since the accident. Since, however, the \(^{137}\)Cs activity levels in mushrooms have remained practically constant (except for course of physical disintegration), their consumption today has a greater relative importance with respect to other routes of ingestion. In a study of the dose levels in different areas of subarctic Russia, Kaduka et al. (2005) concluded that, while in 1987 the consumption of mushrooms only contributed 3.6% of the dietary dose, in the period 1991-2004, this had risen to 52%.

In order to reduce the dose from mushroom consumption in countries with a significant Chernobyl contribution, a recommendation was made to prohibit their consumption in zones where the deposition of \(^{137}\)Cs was either greater than 555 kBq/m\(^2\) or greater than 185 kBq/m\(^2\) and the internal dose was above 1 mSv. Indeed, this restriction of mushroom consumption reduced the dose levels by a factor of 3, and was the best action in terms of the cost-benefit ratio (Jacob et al. 2001; Shaw et al. 2001). However, in countries not highly contaminated by Chernobyl fallout, the annual effective dose due to mushroom ingestion was lower than those

radiocaesium content significantly: cleaning and washing 20%, boiling and salting 40-50%, frying or pickling 70% (Kenigsberg et al. 1996; Beresford et al. 2001; Kalach 2001). Drying was the only mode of preparation that increased the radioactivity content, due to the loss of water. The form of preparation thus affects radiocaesium intake, and should be taken into account to correctly estimate the internal dose. Other radionuclides were also found to have a relatively significant contribution to the effective dose. In the case of \(^{210}\)Po, this was estimated at 8.3 \(\mu\)Sv/year, similar to that of radioactivity in the same ecosystem (Kirchner et al. 1998). The contribution of natural radionuclides to the dose due to consumption of fungi was especially significant in ecosystems that were not highly contaminated, such as the case of Spain. Fig. 4 shows the range of the contribution from natural and anthropogenic radionuclides to the dose due to ingestion of mushrooms in this kind of ecosystems. The radioactivity contribution was greater than that of the other anthropogenic radionuclides considered – \(^{90}\)Sr, \(^{239,240}\)Pu, and \(^{241}\)Am. Nevertheless, in some cases of low radioactivity content, the contribution of natural radionuclides – \(^{40}\)K, \(^{226}\)Ra, and \(^{228,230,232}\)Th – may be similar to or even greater than that of \(^{137}\)Cs.

**CONCLUSIONS**

Mushrooms can bioaccumulate radionuclides in forest ecosystems. Radioactivity has been the most widely and intensively studied radionuclide, showing a great range of variation of about eight orders of magnitude. The affinity of some species of fungi for radioactivity was so high that they accumulated it even 40 years after the deposition, as in the case of global fallout. The accumulation or transfer of other radionuclides has been far less studied, especially the cases of alpha emitters. Generally, their activity levels were lower than that of radioactivity, except in the case of \(^{40}\)K in countries that are not highly contaminated. The activity levels of \(^{40}\)K and radioactivity in mushrooms were uncorrelated, and their frequency distributions were significantly different – log-normal type for radioactivity and Gaussian for \(^{40}\)K. These findings were taken to imply that the content of K in the mushrooms is somehow self-controlled. Short-lived radionuclides such as \(^{125}\)Sb or \(^{106}\)Ru were only detected shortly after the Chernobyl accident. Of the anthropogenic radionuclides that we found reported in the literature, \(^{239,240}\)Pu and \(^{241}\)Am presented the lowest activity levels. With respect to natural radionuclides, their degree of variation was less than that of radioactivity. In particular, the relatively high activity levels of \(^{40}\)K and thorium and uranium isotopes were surprising given their low bioavailability in
soils.

The radioactive content of mushrooms can be influenced by several factors affecting the growth of fungi. The source term had a great influence. In sites greatly affected by the Chernobyl accident, there was a significant rise in their activity. The degree of bioavailability of radionuclides in that layer was key factors in determining the soil-to-fungus transfer of radionuclides and their final accumulation in the fruiting bodies. The content of stable elements in that soil layer also affected the radioactivity of mushrooms which was observed to correlate with stable Rb and Cs in the sense of decreasing 137 Cs content with increasing content of those elements.

Currently, the consumption of mushrooms is the main contribution to the internal dose in highly contaminated areas, mainly because of their high radioactive content. In some arctic areas, there has been observed a steady increase in the importance of the consumption of mushrooms to the dietary intake of radionuclides, due mainly to a decrease of its content in agricultural produce. In other areas with a lower radionuclide contamination, the contribution of other radionuclides may be similar to or even higher than that of radionuclides. The radionuclide content in mushrooms was estimated to be readily digestible by man, so that almost all the radionuclides present in mushrooms contributed to the internal dose. Mushrooms’ radionuclide content, and consequently, the dose, can be reduced by some methods of preparation such as boiling or frying.

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