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## Review Radioactivity in mushrooms: A health hazard?

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#### ABSTRACT

Mushrooms are a complementary foodstuff and considered to be consumed locally. The demand for mushrooms has increased in recent years, and the mushroom trade is becoming global. Mushroom origin is frequently obscured from the consumer. Mushrooms are considered excellent bioindicators of environmental pollution. The accumulation of radionuclides by mushrooms, which are then consumed by humans or livestock, can pose a radiological hazard. Many studies have addressed the radionuclide content in mushrooms, almost exclusively the radiocaesium content. There is a significant lack of data about their content from some of the main producer countries. An exhaustive review was carried out in order to identify which radionuclide might constitute a health hazard, and the factors conditioning it. Regulatory values for the different radionuclides were used. The worldwide range for radiocaesium, <sup>226</sup>Ra, <sup>210</sup>Pb, and <sup>210</sup>Po surpasses those values. Appropriate radiological protection requires that the content of those radionuclides in mushrooms should be monitored.

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#### 1. Introduction

Mushroom consumption, particularly that of wild mushrooms, has traditionally been considered as local. In the case of wild mushrooms, it has generally been considered that they are usually collected and consumed by the local population. This was the key factor in the successful dose reduction in areas with heavy radioactive fallout (Jacob et al., 2001; Shaw, Robinson, Holm, Frissel, & Crick, 2001). However, this local market hypothesis may no longer be valid. The demand for wild edible mushrooms has increased substantially in the last years (Voces, Díaz-Balteiro, & Alfranca,







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2012), and their consumption is considered a delicacy in many countries. This is reflected in a significant rise in the distribution of these products (Pettenella, Secco, & Maso, 2007). According to FAO estimates, the production of mushrooms and truffles increased steadily from  $4.9 \times 10^{11}$  kg in 1961 to a maximum of  $6.4 \times 10^{12}$  kg in 2008 (FAOSTAT, 2012). China was the main producer (more than 70% of the total production) followed by the USA, the Netherlands, Poland, and Spain (FAOSTAT, 2012). The mushroom trade is becoming global. In 2011, Spain's exports and imports to and from other EU countries were  $1.8 \times 10^{10}$  and  $2.4 \times 10^{11}$  kg, respectively, mainly canned and preserved (Cámaras de Comercio, 2012). A negative aspect of trade in mushrooms is the usual lack of transparency of value chain traceability, although some efforts have been made to improve it (Voces et al., 2012). This global market implies that, in general, the consumer is unaware of the product's origin (Voces et al., 2012).

Mushrooms are considered excellent bioindicators of environmental pollution, since they are known to bioaccumulate heavy metals. The necessary estimation of their toxicological risk is often hindered by the lack of available data (Kalač, 2010). Most radionuclides, whether anthropogenic or naturally occurring, are also heavy metals, and can be bioaccumulated by mushrooms. Their contents in mushrooms can pose a health hazard, as has occurred in areas heavily contaminated by radioactive fallout (e.g., those affected by the Chernobyl accident) since they are higher than in other foodstuffs, in particular forest products such as bilberries (Horyna, 1991; IAEA, 2006; Mietelski & Jasinska, 1996; Skuterud, Travnikova, Balonov, Strand, & Howard, 1997). In areas affected by the Chernobyl fallout, the consumption of wild mushrooms led to increased body content of radiocaesium in the population, which also showed a seasonal trend linked to the fructification of mushrooms, being higher in autumn (Shutov et al., 1996; Skuterud et al., 1997). The initial recommendation to prohibit their consumption was one of the most successful actions reducing the received dose by the population in areas with significant Chernobyl fallout (Jacob et al., 2001; Shaw et al., 2001). Likewise, the consumption and distribution of log-cultivated shiitake mushrooms (Lentinula edodes) were restricted in areas affected by the Fukushima accident (MAFF, 2012). These restraints were lifted when the mushrooms' radioactive contents fell below regulatory values (MAFF, 2012; Hamada & Ogino, 2012).

Another indirect pathway which can affect the human population, especially critical groups in the Arctic, is the consumption of game (Strand et al., 2002). Reindeer and caribou eat mushrooms whenever available. Indeed, a rumen content of up to 20% of mushrooms has been reported in an individual moose (Johanson, Bergström, Eriksson, & Erixon, 1994). Abundant spores of *Xerocomus* spp. and *Hypholoma capnoides* fungi and increased <sup>137</sup>Cs content have been observed in the faeces of roe deer (Strandberg, 1994a). In some countries, this lead to a significant, highly seasonal, enhancement of the radiocaesium content in game meat, with maxima in the second half of the year (Hove, Pedersen, Garmo, Solheim, & Staaland, 1990; Zibold, Drissner, Kaminski, Klemt, & Miller, 2001).

The analysis of the radioactive content of mushrooms has mainly focused on radiocaesium. There are essentially two reasons for this. One is that it is a long-lived anthropogenic radionuclide ( $T_{\nu_2}$  = 30.2 y) that has been released into the environment by atmospheric nuclear weapons tests and various accidents involving nuclear materials (UNSCEAR, 2000). The second is that it is a chemical analogue of potassium. Other anthropogenic ( $^{90}$ Sr,  $^{239+240}$ Pu,  $^{241}$ Am, etc.) and naturally occurring ( $^{40}$ K, and members of the natural uranium and thorium series, among others) radionuclides have been less studied, even though their content and radiological impact can on occasions surpass those of radiocaesium (Guillén, Baeza, Ontalba, & Míguez, 2009; Vaarama, Solatie, & Aro, 2009).

The main objective of the present work was to estimate the anthropogenic and naturally occurring radionuclide content of mushrooms, in order to assess whether they may pose a health hazard. This content depends on several parameters, including the level of radioactive fallout or naturally occurring radionuclides in the environment, the mushroom species, its nutritional mechanism, mycelium depth, climate, bioavailability of the radionuclide, etc. According to estimates made mainly from the radiocaesium content, its contribution to the internal dose may be significant in some producing areas. However, other radionuclides with greater radiotoxicity, in particular <sup>210</sup>Pb and <sup>210</sup>Po, can also contribute to the internal dose (Guillén, Baeza, Ontalba, et al., 2009): Vaarama et al., 2009). The current legislation is also reviewed in order to assure the correct protection of the population, which is especially important taking into account the global market.

#### 2. Radionuclide content in mushroom

Table 1 lists the range of anthropogenic and naturally occurring radionuclides in mushrooms collected worldwide as reported in the literature. These determinations of the radioactive content of mushrooms have focused mainly on anthropogenic radionuclides as biomonitors of the radiological status of the environment. Among them, radiocaesium is the most analysed in mushrooms, mainly because of its environmental significance. The determination of other  $\alpha$ - and/or  $\beta$ -emitter contents in mushrooms has been less frequent, reflecting in part the long and costly radiochemical procedures associated with their determination, even though these radionuclides can also be major contributors to the internal dose.

#### 2.1. Anthropogenic radionuclides

#### 2.1.1. Radiocaesium

The range of variation of radiocaesium content in mushrooms worldwide after 1986 is huge – about nine orders of magnitude (Table 1). Table 2 summarizes the range of <sup>137</sup>Cs (expressed as Bg/kg d.w.) reported in the literature for many countries, and the species which presented the highest activity levels. Data from the Fukushima accident were not included in the table since they were not as yet readily available in the literature. Also noteworthy is the almost complete absence of data, or at least of readily available data in the literature, from China, the world's principal mushroom producer, as well as from other non-European countries among the top 20 producers (FAOSTAT, 2012). As well as <sup>137</sup>Cs, <sup>134</sup>Cs was also released into the environment. However, this radionuclide is not included in Table 2, because it has a shorter half-life  $(T_{1/2} = 2.06 \text{ y})$  than <sup>137</sup>Cs  $(T_{1/2} = 30.1 \text{ y})$ , and the <sup>134</sup>Cs/<sup>137</sup>Cs ratio is indicative of the different releases that have occurred. Indeed, <sup>134</sup>Cs has only been reported for countries affected by a relative recent deposition of radionuclides, such as Chernobyl or Fukushima, while it is absent from those affected by older depositions, such as global fallout in the case of Spain.

There are several factors affecting the radiocaesium content in mushrooms. First, the quantity deposited onto soil is closely related to the range of the contents, especially to the maximum content. The countries seriously affected by the Chernobyl fallout (UNSCEAR, 2000) presented the highest content. The Chernobyl fallout was inhomogeneous all over the countries affected. Therefore, areas with different radioactive contamination can be found within a given country (Mietelski, Jasinska, Kozak, & Ochab, 1996; UNSCEAR, 2000). If mushrooms collected within the same local area or forest are considered, the range of variation is reduced to 1–3 orders of magnitude (Gentili, Grenigini, & Sabbatini, 1991; Mietelski, Jasinska, Kubica, Kozak, & Macharski, 1994; Mietelski, Dubchak, Blazec, Anielska, & Turnau, 2010). The maximum content

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