Artificial and natural radioactivity in edible mushrooms from Sao Paulo, Brazil

Journal of Environmental Radioactivity, n. 113, p. 150-154, 2012
http://www.producao.usp.br/handle/BDPI/48898

Downloaded from: Biblioteca Digital da Produção Intelectual - BDPI, Universidade de São Paulo
Artificial and natural radioactivity in edible mushrooms from Sao Paulo, Brazil

L.P. de Castro a, V.A. Maihara a,*, P.S.C. Silva a, R.C.L. Figueira b

a Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP), Av.Prof. Lineu Prestes 2242, CEP 05508-000 Sao Paulo, Brazil
b Instituto Oceanográfico da Universidade de Sao Paulo, Pça. do Oceanográfico, 191, CEP 05508-120 Sao Paulo, Brazil

Abstract

Environmental biomonitoring has demonstrated that organisms such as crustaceans, fish and mushrooms are useful to evaluate and monitor both ecosystem contamination and quality. Particularly, some mushroom species have a high capacity to retain radionuclides and some toxic elements from the soil and the air. The potential of mushrooms to accumulate radionuclides in their fruit-bodies has been well documented. However, there are no studies that determine natural and artificial radionuclide composition in edible mushrooms, in Brazil. Artificial (137Cs) and natural radioactivity (40K, 226Ra, 228Ra) were determined in 17 mushroom samples from 3 commercialized edible mushroom species. The edible mushrooms collected were Agaricus sp., Pleurotus sp. and Lentinula sp. species. The activity measurements were carried out by gamma spectrometry. The levels of 137Cs varied from 1.45 ± 0.04 to 10.6 ± 0.3 Bq kg⁻¹, 40K levels varied from 461 ± 2 to 1535 ± 10 Bq kg⁻¹, 226Ra levels varied from 14 ± 3 to 66 ± 12 Bq kg⁻¹ and 228Ra levels varied from 6.2 ± 0.2 to 54.2 ± 1.7 Bq kg⁻¹. 137Cs levels in Brazilian mushrooms are in accordance with the radioactive fallout in the Southern Hemisphere. The artificial and natural activities determined in this study were found to be below the maximum permissible levels as established by national legislation. Thus, these mushroom species can be normally consumed by the population without any apparent risks to human health.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction

Artificial and natural radionuclides are found in diverse environmental compartments such as oceans, rivers, soils, rocks, vegetables and animal as well as human body tissues (Hu et al., 2010). Therefore, human beings and their environment are continuously exposes to these types of radiation, of which 81% can be attributed to natural radiation. The other 19% comes from artificial sources (Mazzilli et al., 2002).

Food is one of the main sources for elements and radionuclides for humans. This being true, radioactivity measurements in the environment and foodstuffs have become very important to evaluate the radiation levels to which man is exposed to either directly or indirectly.

Environmental biomonitoring has demonstrated that organisms such as crustaceans, fish and mushrooms are useful to evaluate and monitor both ecosystem contamination and quality (Marzano et al., 2001). Of these organisms, mushrooms stand out as they are excellent nutritional sources of proteins, fibers, vitamins and minerals, such as K, P and Fe and present low Na concentrations. Presently, there are over 2000 known species, many of which are edible. However, some species are highly toxic and poisonous if consumed can lead to death (Keizer, 2001).

According to Kuwahara et al. (2005) and Bazala et al. (2008), different mushroom species have the capacity to retain high concentrations of radionuclides and metals from the soil. These authors state that mushroom’s longevity and wide mycelium make the mushroom an excellent environmental bioindicator. Among those radionuclides that can be accumulate by mushrooms are 40K, 137Cs, 226Ra, 228Ra and 228Ra.

Thus, the bioaccumulation characteristics of mushrooms can be used to detect even very low contamination levels. After the Chernobyl event, mushrooms have been used by many researchers as important biomonitor for high artificial radionuclide contamination. Battiston et al. (1989) analyzed the Clitocybe infundibuliformis, Cantharellus lutescens e Boletus cavipes edible mushroom species and found 137Cs concentrations from 95 to 27,626 Bq kg⁻¹. According to these researchers, these high 137Cs levels were not only related to soil contamination, but also due to the fact that some mushroom species presented high affinity to concentrate some radionuclides.

Kammerer et al. (1994) determined 137Cs level in 83 wild mushroom species in three different regions of Germany. The levels varied from 2 to 15,000 Bq kg⁻¹ and the Xerocomus badius species presented the highest 137Cs level.
2. Materials and methods

2.1. Sampling

The edible mushroom samples were collected from different commercial points in São Paulo metropolitan region, specifically in Municipal Markets. Some samples were collected directly from producers located in the cities of Mogi das Cruzes, Mirandópolis, Suzano and Juquitiba.

About 20 g of each mushroom sample were placed in polyethylene bottles previously cleaned with 10% nitric acid (Merck, Darmstadt, Germany) and Milli-Q water. The bottles (65 mm in diameter and 19 mm of height) were stored for approximately 35 days until the respective counting.

2.2. Sample preparation

All samples were cut in small pieces with a plastic knife and put in petri plates with Ti blades.

2.3. Gamma spectrometry

About 20 g of each freeze-dried mushroom sample were placed in a polyethylene bottle previously cleaned with 10% nitric acid (Merck, Darmstadt, Germany) and Milli-Q water. The bottles (65 mm in diameter and 19 mm of height) were stored for approximately 35 days until the respective counting. The activities of gamma emitters were determined by using a High purity Ge (HPGe) POP TOP model detector (EG&G ORTEC, Oak Ridge, USA) with 1.9 keV resolution for the 1332.2 keV of $^{56}$Co and 50% relative efficiency.

2.4. $^{137}$Cs, $^{40}$K, $^{228}$Ra and $^{226}$Ra determination

To determine the low $^{137}$Cs levels the methodology developed by Figueira et al. (2007) was applied. Consequently, the same methodology was used for $^{40}$K, $^{228}$Ra and $^{226}$Ra determinations. $^{137}$Cs (661.6 keV) and $^{40}$K (1460.8 keV) were determined directly by their own photopeaks. In the case of $^{226}$Ra and $^{228}$Ra, both were determined through their daughter radionuclides: $^{214}$Bi (609.3 keV) and $^{228}$Ac (911.2 keV), respectively.

The adopted procedure consisted, initially, in an accumulating counting of background radiation (BG) measuring one empty plastic bottle in intervals of 25,000 s, from 100,000 to 200,000 s. Later, reference materials that were placed in the same geometric bottles and stored up to 20 days, were submitted to the same counting procedure for the BG, in order to calculate the detector efficiency.

Linear regression fits were made for the activity by counting time (BG and reference materials). Then, the detector efficiency was determined by Eq. (1):

$$ \varepsilon = \frac{\alpha_{RM} \cdot a_{BG}}{m_{RM} \cdot \alpha_{RM}} $$

where $\varepsilon$ is the efficiency for the radionuclide determined, $\alpha_{RM}$: $a_{BG}$ is the angular coefficient of the regression curve for the reference material and BG; $m_{RM}$ is the reference material mass, in kilograms; $\alpha_{RM}$ is the reference material activity concentration (Bq kg$^{-1}$), adjusted to the date of the analysis.

The same accumulative method used for the reference materials was used for the sample counting. The activity concentrations were determined from Eq. (2):

$$ A = \frac{a_{S} \cdot a_{BG}}{m_{A} \times t \times \varepsilon} $$

$A$ is the sample activity concentration in Bq kg$^{-1}$; $a_{S}$, $a_{BG}$ is the angular coefficient of the regression curve for the sample and BG; $m_{A}$ is the sample mass in kg; $t$ is the counting time in seconds; $\varepsilon$ is the efficiency for the radionuclide determined.

3. Results and discussion

The detector efficiency ($\varepsilon$) was obtained from measurements of reference materials: IAEA-300 (sediment marine), for $^{137}$Cs and $^{40}$K and IAEA-300 (sediment marine) and IAEA-327 (soil) for $^{228}$Ra and $^{226}$Ra. The reference material data used for the efficiency determination and the efficiency values obtained are presented in Tables 1 and 2.

For the validation of the methodology IAEA-307 (sea plant), IAEA-414 (mixed fish), IAEA-375 (soil) and IAEA — Mushroom reference materials were analyzed. The mushroom reference material was prepared within the frame of an IAEA Intergovernmental Technical Cooperation Project (Waheed et al., 2007; Polkowski-Motrenko and Rossbach, 2007). The results are presented in...
as depth of mycelium and substrate cultivation. However, according to Martine and Ramsey (2008) to evaluate the activity concentrations in the edible mushroom species analyzed varied from 14 to 66 Bq kg$^{-1}$ (dry weight). According to Gillet and Crout (2000), different factors determine the $^{137}$Cs levels in mushroom as well as the ability of some species to retain this radionuclide such as depth of mycelium and substrate cultivation.

The activity concentration of $^{137}$Cs varied from 1.45 ± 0.04 to 10.6 ± 0.3 Bq kg$^{-1}$ (dry weight). According to Gillet and Crout (2000), different factors determine the $^{137}$Cs levels in mushroom as well as the ability of some species to retain this radionuclide such as depth of mycelium and substrate cultivation.

The $^{137}$Cs levels determined in the edible mushroom species were lower than the reported levels by Teherani (1988) in Austria that analyzed several European mushroom species a year after the Chernobyl accident. The levels determined varied from 18 to 1535 ± 10 Bq kg$^{-1}$ (dry weight). On the other hand, unlike $^{137}$Cs, $^{40}$K activity levels were comparable with other studies. Wang et al. (1998), in Taiwan, found levels ranging from <50 to 1230 Bq kg$^{-1}$ and Malinowska et al. (2006) in Poland reported values between 180 and 1520 Bq kg$^{-1}$ (dry weight). These studies seem to suggest that $^{40}$K incorporation is self-regulated by fungi, whereas $^{137}$Cs appears not to be regulated by fungi. Kuwahara et al. (2005) proposed that the $^{40}$K variations in mushroom are related to local geology, cultivation methods, use of fertilization, culture age, among other factors.

### Table 1

<table>
<thead>
<tr>
<th>Reference material</th>
<th>Reference date for decay correction</th>
<th>$^{137}$Cs</th>
<th>$^{40}$K</th>
<th>Efficiency values (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Marine sediment IAEA 300</td>
<td>01/01/1993</td>
<td>1066.6 (1046–1080)</td>
<td>1059 (1046–1226)</td>
<td>1.90 0.19</td>
</tr>
</tbody>
</table>

### Table 2

<table>
<thead>
<tr>
<th>Reference material</th>
<th>Reference date for decay correction</th>
<th>$^{226}$Ra</th>
<th>$^{228}$Ra</th>
<th>Efficiency values (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Marine sediment IAEA 300</td>
<td>01/01/1993</td>
<td>61.6 (59.0–63.9)</td>
<td>56.6 (54.4–60.2)</td>
<td>0.73 1.19</td>
</tr>
<tr>
<td>Soil IAEA 327</td>
<td>31/12/1994</td>
<td>38.7 (37.8–39.6)</td>
<td>35.1 (32.7–35.5)</td>
<td>0.76 1.56</td>
</tr>
</tbody>
</table>

### Table 3

The activity concentration for $^{137}$Cs obtained was 10.6 ± 0.3 Bq kg$^{-1}$ which was lower than the allowed level by Brazilian legislation (CNEN-301/006), which is 1000 Bq kg$^{-1}$ for foodstuffs.

In regards to $^{40}$K in Brazilian edible mushrooms, all species presented the activity concentrations for this radionuclide and the levels varied from 461 ± 2 to 1535 ± 10 Bq kg$^{-1}$ (dry weight).

The maximum value for $^{137}$Cs was reported by Baeza et al. (2005) in Spain whose values varied from 140 to 1520 Bq kg$^{-1}$ (dry weight). For comparison purposes the concentrations of Ra-isotopes in vegetables in general found in literature varied from 0.6 ± 0.3 to 17 ± 7 Bq kg$^{-1}$ for $^{228}$Ra and 0.3 ± 0.04 to 17 ± 5 Bq kg$^{-1}$ for $^{226}$Ra according to Shanthi et al. (2010) in Indian vegetables, fruits and tuber. Pietrzak-Flis et al. (2001) found lower and higher activity concentrations in the range of 28–93 Bq kg$^{-1}$ for $^{226}$Ra and 36–117 Bq kg$^{-1}$ for $^{228}$Ra.

All the activity concentrations measured for Ra-isotopes in Brazilian mushrooms lie within the range of vegetable foodstuffs as follows:

### Table 4

<table>
<thead>
<tr>
<th>Reference material</th>
<th>$^{40}$K (Bq kg$^{-1}$)</th>
<th>$^{137}$Cs (Bq kg$^{-1}$)</th>
<th>$^{226}$Ra (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IAEA Mushroom</td>
<td>962 ± 4 (15.3)$^a$</td>
<td>1136 (1046–1226)$^b$</td>
<td>–</td>
</tr>
<tr>
<td>IAEA-414</td>
<td>484 ± 2 (0.7)$^a$</td>
<td>480 (461–498)$^b$</td>
<td>–</td>
</tr>
<tr>
<td>IAEA-307</td>
<td>–</td>
<td>5.5 ± 0.2 (12.5)$^a$</td>
<td>–</td>
</tr>
<tr>
<td>IAEA-375</td>
<td>–</td>
<td>–</td>
<td>23.7 ± 4.4 (15.6)$^a$</td>
</tr>
</tbody>
</table>

The reference materials used do not possess certified values for $^{228}$Ra.

$^a$ RE – Relative error.

$^b$ 95% Confidence interval.
reported in literature. According to UNSCEAR (2000) reference values for $^{226}$Ra and $^{228}$Ra are 0.05 and 0.04 Bq kg$^{-1}$ for leafy vegetables and 0.03 and 0.02 Bq kg$^{-1}$ for root vegetables and fruits. The activity concentrations vary in a range from 0.002 to 1.150 Bq kg$^{-1}$ for $^{226}$Ra and from 0.11 to 0.22 Bq kg$^{-1}$ for $^{228}$Ra in the same products.

There are many factors affecting the level of radioactivity in mushrooms. Some of these include soil, pH and species of mushrooms and other groups. As it is known, fertilizers can contain different radionuclides. This same heterogeneity is also observed in relation to the sample origin. In this case the variation obtained can be attributed directly to the form of cultivation, relating to the use of certain fertilizers that can significantly influence the radioactive contents. As it is known, fertilizers can contain different radionuclides in their composition (Luca et al., 2009; Bituh et al., 2009).

4. Conclusion

The gamma spectrometry methodology allowed to determining the $^{137}$Cs, $^{40}$K, $^{226}$Ra and $^{228}$Ra levels present in edible mushroom samples with good precision and accuracy. The $^{137}$Cs levels in the edible mushrooms are in accordance with the Southern Hemisphere fallout level. The highest value found in this study for the activity of $^{137}$Cs was very low in comparison with the maximum allowed by national legislation for foodstuffs. $^{40}$K levels obtained are in agreement with values given in other studies. Thus, the edible mushrooms analyzed in this study do not present a risk to their consumers, even though consumption of this type of foodstuff is still small in Brazil. However, it is always necessary to establish controls not only the concentration of $^{137}$Cs in mushrooms, but also in other foods such as meat, milk, fruit, etc. In the case of $^{40}$K, which is evenly distributed in the body and is under homeostatic control, is less dangerous to human health than $^{137}$Cs. The $^{226}$Ra and $^{228}$Ra levels obtained were also comparable with literature values.

References


Martine, C.D., Ramsey, M.L., 2008. Accumulation of radiocesium by mushrooms in


