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Radionuclides content in *Xerocomus badius* and other commercial mushrooms from several regions of Poland

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Abstract

In this study the levels of ^{137}Cs, mostly originating from the Chernobyl fallout, as well as levels of natural radionuclides ^{40}K and ^{210}Pb present in caps and stalks of edible mushroom samples and soil from Poland were measured. The analytical technique used was alpha spectrometry to determine ^{210}Pb through its granddaughter ^{210}Po. The ^{137}Cs and ^{40}K activity concentrations were determined by gamma spectrometry. The radionuclide activities in *Xerocomus badius* were in the ranges: 330–6670 (^{137}Cs), 180–1520 (^{40}K) and 0.70–32.0 (^{210}Pb) Bq kg^{-1} d.w. The highest measured concentration of ^{137}Cs was in the caps of *X. badius* from the Borecka Forest. The caps of examined mushrooms showed significantly higher activity concentrations compared to the stalks. In order to estimate the degree of accumulation of each element by mushrooms, transfer factor (TF) soil–cap and soil–stalk values were calculated. Also, some commercial mushrooms were examined such as dried, frozen and marinated products.

Keywords: Radionuclides; Edible mushrooms; Soil; Transfer factor

1. Introduction

In forest ecosystems, radionuclides are deposited in surface organic layers onto the trees and other plants like grass, berries moss and lichen. In the case of ^{40}K and ^{210}Pb, they are also consubstantial with the soils of forest ecosystems. Mushrooms, due to their ability to accumulate caesium isotopes, represent an important pathway to the food chain for radioactivity arising from radioactive fallout after nuclear weapons tests and from the Chernobyl accident (Steiner, Linkov, & Yoshida, 2002; Yoshida & Muramatsu, 1994).

The behaviour and transport of radionuclides in soil, plants and mushrooms have been the objects of many investigations (Gaso et al., 2000; Grabowski et al., 1994; Korky & Kowalski, 1989; Vera Tome, Blanco Rodriguez, & Lozano, 2003). Since macrofungi are an integral part of forest ecosystems, the soil-to-mycelium transfer of radionuclides and the relationships between mycelium and symbiotic plant species regarding element absorption and translocation represent an important part of radioecological studies. It has been suggested by Gillett and Crout (2000) that the main factors controlling the ^{137}Cs levels in mushrooms are the depth of the mycelium and the ecophysiological behaviour of the fungi. Various species of macrofungi accumulate radionuclides to various degrees. *Xerocomus badius* is known to be a good accumulator of radiocaesium because of presence of a dye in the caps's skin that complexes caesium and potassium (Aumann, Clooth, Steffan, & Steglich, 1989).

Radiocaesium is of particular concern in the natural environment because of a long half-life period (30.1 years), easy migration in the trophic chains and great bioavailability. In addition, characteristic soil properties

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of surface soil horizons in forests such as low pH value, low clay content and high organic carbon content, enhance radiocaesium bioavailability (Kruyts & Delvaux, 2002; Thiry, Kruyts, & Delvaux, 2000) and may promote its uptake by mushrooms (Niesiobędzka, 2000).

The aim of the present investigation was to determine the levels of an anthropogenic radioisotope ^{137}Cs and the natural radioisotopes ^{40}K and ^{210}Pb in *X. badius* and underlying soil samples from forested areas and in other commercial mushrooms *X. badius*, *Boletus edulis* and *Laccinum scabrum* dried, frozen and marinated, available in local market, and to assess the risk for humans.

2. Materials and methods

Mushrooms were collected in years 1996–1998 in various forests located mostly in north- and northeastern parts of Poland. The sampling sites are shown in Fig. 1. Each specimen was thoroughly cleaned to remove external contamination and debris such as moss, tree leaves and needles, and soil particles. Mushrooms were divided into caps and stalks, and each part was analyzed separately. The dried material was ground in an agate mortar and thoroughly mixed to obtain a homogenous sample. For each sampling location, a composite sample was made of 7–16 specimens of approximately the same size within the species. Commercial mushrooms, available in dried and marinated forms, were analyzed in their original form. The total number of mushroom samples analyzed was 132. Thirty one soil samples, were collected from the same sites as for the mushrooms. The samples were taken from the uppermost 10 cm soil layer. Coarse objects such as stones, tree leaves and branches, worms and insects etc., were removed by hand-picking. The remaining material was sieved through a 1 mm plastic sieve and the part that passed through was retained for analysis.

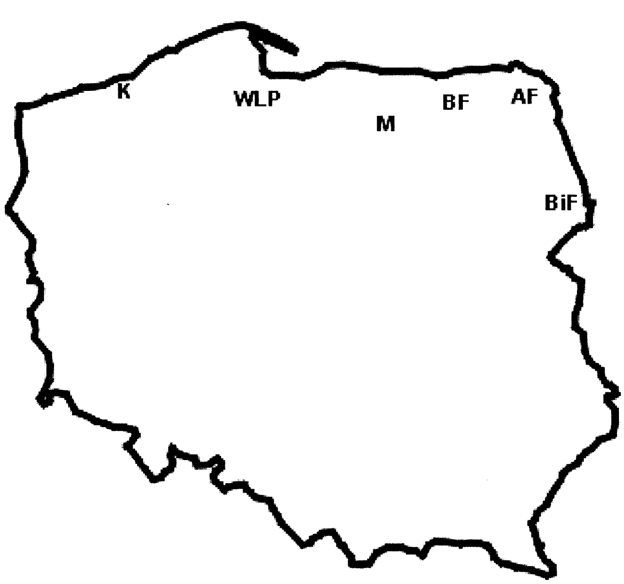

Fig. 1. Location of sampling area (K – adjacent area of Kolobrzeg, WLP – Wdzydzki Landscape Park, M – adjacent area of Morag, BF – Borecka Forest, AF – Augustowska Forest, BiF – Bialowieska Forest).

Mushroom and soil subsamples were also analysed for concentrations of trace elements and macroelements. The data obtained have been reported elsewhere (Malinowska, Szefer, & Falandysz, 2004).

The gamma-ray emitting radionuclides ^{137}Cs and ^{40}K were measured by gamma spectrometry while ^{210}Pb was determined via its granddaughter ^{210}Po by alpha spectrometry. For gamma-spectrometric measurements, the dry material was placed in a cylindrical container of diameter 60 mm. The filling height varied from 3 to 20 mm, depending on the amount of material available, which was equivalent to 5–30 g of dry matter. The apparatus consisted of a high purity n-type germanium detector with 1.8 KeV resolution at 1.33 MeV ^{60}Co line, and 20% relative efficiency, coupled to a Canberra Workstation S 100. The samples were counted for 2–3 days each. Minimum detectable activity concentrations were calculated according to Currie's formula (Currie, 1968):

$$\mathrm{MDA} = \frac{2.71 + 4.66\sqrt{B}}{E \cdot t \cdot M},$$

where B is count number of the blank, t is counting time (s), M is sample weight (kg) and E – calibration factor (counts/Bq) dependent on the radionuclide and the sample geometry. The E values for each filling height were established experimentally using a reference material IAEA-373 (grass). The values of minimum detectable activity (MDAs) were found to be between 0.1 and 0.5 Bq kg^{-1} for ^{137}Cs and 20–30 Bq kg^{-1} for ^{40}K. All ^{137}Cs data were decay-corrected to the same day of sampling.

Polonium-210 was determined in the same material as used for gamma spectrometry. Approximately 0.5 g of dry matter was wet-digested in an Automatic Microwave Digestion System (MLS 1200) with nitric acid in the presence of calibrated ^{209}Po solution as yield monitor. The polonium was deposited on silver discs and measured for 1–3 days in an alpha particle spectrometer equipped with PIPS detectors. The MDA for ^{210}Po was estimated at 0.1–0.3 Bq kg^{-1} d.w.

Since the time lapse between samples collection and analysis was two years or more, the ^{210}Po–^{210}Pb pair was assumed to be in secular equilibrium. Decay correction to the sampling time was, therefore, made using the ^{210}Pb half-life of 22.26 years.

3. Results and discussion

3.1. General

The results of analyses are shown in Tables 1 and 2. They are expressed on a dry weight basis and decay-corrected to sampling dates. The error terms represent expanded standard uncertainties (2k). They comprise both counting and calibration errors, added according to the error propagation rule.

3.2. Radionuclide activity concentrations in X. badius and other commercial mushrooms

The highest ^{137}Cs activity concentration was found in the caps of *X. badius*, amounting to 6670 Bq kg^{-1}. The measured ^{137}Cs concentrations of the various species are in good agreement with those reported elsewhere for Poland (Mietelski, Jasińska, Kubicka, Kozak, & Macharski, 1994). In that study, relatively low levels were found in *B. edulis* and *L. scabrum* while higher levels were found in *X. badius*. Mushrooms collected in coniferous forests were characterised by higher ^{137}Cs activity than those from deciduous forests (Heinrich, 1992). It seems to be associated with more available Cs in thick humus and therefore with greater Cs uptake (Kruyts & Delvaux, 2002; Thiry et al., 2000). Also, leaching from dropped needles and bark radiocaesium is accumulated in the top layer and can increase ^{137}Cs activity in growing mushrooms. The highest activity concentration of ^{137}Cs in commercial mushrooms was found in dried slices of fruiting bodies of *X. badius* – 4290 Bq kg^{-1}.

The ^{40}K concentration values in the caps and stalks of *X. badius* ranged from 498 to 1480 Bq kg^{-1} (average value of 990 Bq kg^{-1}) and from 180 to 1520 Bq kg^{-1} (average value of 910 Bq kg^{-1}), respectively. These results are similar to those reported for mushrooms collected in different forest areas (Mietelski et al., 1994; Yoshida & Muramatsu, 1994). According to Byrne (1988), ^{40}K is present in mushrooms in an almost constant concentration, whereas ^{137}Cs or ^{210}Pb contents in fruiting bodies vary widely. It seems to indicate that the incorporation of stable potassium, and therefore of ^{40}K is self-regulated by the fungus's own physiological requirements. In opposition to ^{40}K, the content of ^{137}Cs is not self-regulated by the fungus (Baeza et al., 2004).

In the analysed market mushrooms, activities of natural isotope ^{40}K were from 450 Bq kg^{-1} in whole fruiting bodies of *B. edulis* to 1420 Bq kg^{-1} in the caps of *X. badius*. The values obtained in the present work for ^{40}K in *X. badius* agree with the data reported by other authors (Gaso et al., 1998; Kalač, 2001). The highest average concentration of ^{210}Pb was found in the caps and whole fruiting bodies of *B. edulis*, i.e. 58.9 and 36.6 Bq kg^{-1}, respectively. Kirchner and Daillant (1998) measured activity concentrations of ^{210}Pb in the range 1.76–36.5 Bq kg^{-1} in various mushrooms collected in France.

3.3. Radionuclide activity concentrations in the adjacent soil

Results of the activity concentrations measured in the soil samples are given in Table 1. Contamination by ^{137}Cs of the soils examined was in the range of 11–260 Bq kg^{-1} with the mean content 56.2 Bq kg^{-1}. The highest activity was determined in the Borecka Forest. Activities of ^{40}K in all soil samples were generally

Table 1
Radionuclides activity in the fruiting bodies of bay bolete, *Xerocomus badius* (Fr.) Kühn. ex Gilb., and in the underlying substrate from selected forest areas of Poland [*N* – number of pooled mushroom samples (total number of specimens)]

Sample location	Average activity concentration Bq kg^{-1} d.w. ± standard deviation (min–max)								
	^{137}Cs			^{40}K			^{210}Pb		
	Cap	Stalk	Soil	Cap	Stalk	Soil	Cap	Stalk	Soil
Augustowska Forest *N* = 4 (16)	3960 ± 106 (3270–4440)	3500 ± 87 (1690–4270)	42 ± 4 (26–65)	1120 ± 420 (660–1400)	910 ± 390 (600–1170)	350 ± 46 (270–430)	3.2 ± 0.7 (1.6–4.2)	2.0 ± 0.6 (1.3–2.9)	39 ± 2.3 (33–50)
Bialowieska Forest *N* = 4 (11)	2740 ± 110 (1990–3510)	1860 ± 82 (1240–2770)	104 ± 4 (54–180)	950 ± 590 (498–1370)	730 ± 410 (280–940)	330 ± 41 (320–360)	1.8 ± 0.5 (0.7–2.5)	3.1 ± 0.7 (1.7–7.2)	39 ± 2.0 (27–51)
Borecka Forest *N* = 7 (15)	3160 ± 68 (330–6670)	2620 ± 86 (580–4360)	85 ± 4 (11–260)	1100 ± 260 (870–1250)	890 ± 390 (460–1140)	350 ± 44 (320–400)	3.8 ± 0.8 (1.1–7.2)	16 ± 1.8 (9.0–32)	24 ± 1.8 (9.4–39)
Adjacent area of Morag *N* = 4 (16)	2470 ± 58 (470–5080)	1750 ± 110 (190–3400)	36 ± 2 (15–72)	1070 ± 270 (560–1480)	470 ± 220 (180–790)	240 ± 27 (210–300)	7.0 ± 0.9 (2.5–12)	13 ± 1.5 (6.4–17)	26 ± 1.9 (13–36)
Wdzydzki Landscape Park *N* = 5 (15)	4600 ± 61 (4260–5400)	2180 ± 190 (1390–3620)	34 ± 3 (26–43)	1000 ± 170 (800–1380)	1260 ± 660 (890–1520)	226 ± 40 (220–240)	8.9 ± 0.7 (5.5–11)	4.6 ± 0.7 (1.8–6.1)	25 ± 1.8 (17–31)
Adjacent area of Kolobrzeg *N* = 4 (7)	2300 ± 83 (1430–3110)	2040 ± 72 (1590–2500)	36 ± 3 (21–44)	700 ± 320 (520–850)	1200 ± 380 (1030–1450)	234 ± 32 (180–300)	2.9 ± 0.6 (1.1–5.6)	10 ± 1.0 (0.9–23)	27 ± 1.1 (14–39)

Table 2
Radionuclides activity [Bq kg^{-1}] dry wt., average ± standard deviation in the fruiting bodies (C – cap, S – stalk, O – whole fruiting body) of commercial mushrooms (*N* – number of pooled samples)

Name of species, producer	*N*		^{137}Cs	^{40}K	^{210}Pb
Dried mushrooms					
Boletus edulis slices. Runoland	12	C	296 ± 14	990 ± 97	12.4 ± 0.6
		S	280 ± 12	590 ± 93	10.5 ± 1.1
Xerocomus badius slices. Runoland	12	C	2760 ± 260	1240 ± 87	14.8 ± 0.9
		S	1520 ± 249	897 ± 97	1.11 ± 0.5
Xerocomus badius slices. Borowikowo	12	C	4290 ± 344	966 ± 86	3.24 ± 0.6
Xerocomus badius. IMBA	10	C	1470 ± 117	1020 ± 135	13.2 ± 0.7
		S	1220 ± 98	1130 ± 145	1.97 ± 0.4
Leccinum scabrum. IMBA	10	C	150 ± 6	960 ± 70	10.1 ± 0.7
		S	110 ± 11	695 ± 132	9.22 ± 1.1
Xerocomus badius. Poniatowo	8	C	3810 ± 351	840 ± 83	36.4 ± 1.4
		S	1070 ± 220	460 ± 75	26.2 ± 1.5
Frozen mushrooms					
Boletus edulis. Noris	3	C	900 ± 21	730 ± 107	58.9 ± 3.3
Boletus edulis. Noris	3	O	380 ± 20	450 ± 27	36.6 ± 2.2
Xerocomus badius. Noris	3	C	1900 ± 220	1420 ± 91	4.94 ± 1.0
Marinated mushrooms					
Xerocomus badius. Runoland	3	C	1830 ± 211	1020 ± 84	4.15 ± 1.0

similar, ranging from 180 Bq kg^{-1} in the adjacent area of Kolobrzeg to 430 Bq kg^{-1} in Augustowska Forest. The measured activities of ^{210}Pb in the soil samples were in the range of 9.40–51 Bq kg^{-1}. The highest average concentration of 39 Bq kg^{-1} was found in soil from the Augustowska and Bialowieska Forests. The results of ^{210}Pb activity in soil presented by Kirchner and Daillant (1998) are greater than those estimated in the present study. The main source of ^{210}Pb in surface soil is from atmospheric deposition as well as ^{222}Rn soluble in soil pore water which subsequently decays into ^{210}Pb.

3.4. Radionuclides cap to stalk quotients

In order to indicate differences in the accumulative abilities of radionuclides in particular morphological parts of the fruiting body of *X. badius*, cap to stalk nuclide ratios were estimated and presented in Table 3. It is observed that the caps exhibit higher ^{137}Cs and ^{40}K activity than the stalks. The ratios for ^{137}Cs were greater than unity, ranging from 0.2 to 3.1. The average of the cap to stalk ratios for ^{40}K and ^{210}Pb were 1.4 and 1.1, respectively.

3.5. Transfer factors (TF)

The concept of transfer factor is applied to quantify the translocation and uptake of stable elements and their radioactive isotopes from soil to green plants and fungal fruit bodies (Steiner et al., 2002). The radionuclide transfer factor (TF) from soil to the each part of fruiting body of *X. badius* was calculated according to the formula:

$$TF = \frac{A_{C,S}}{A_{SO}},$$

where $A_{C,S}$ is the activity concentration in cap (c) or stalk (s) and A_{SO} is the activity concentration in the soil (so) on a dry weight basis. The calculated TFs are summarized in Table 3. The average cap–soil and stalk–soil TFs for ^{137}Cs from all the regions studied were in range 34–137 and 21–89, respectively. Accumulation of ^{137}Cs in caps was higher as compared to that of stalks. Mietelski and Jasińska (1996) obtained transfer factor values from soil to a whole fruiting body of *X. badius* in range of 0.7–550 with the average value of 37.7. According to Gaso et al. (1998) accumulation of ^{137}Cs is proportional to its activity in the soil solution.

The average TFs for ^{40}K from underlying soil to the caps and stalks of *X. badius* ranged between 2.9 to 4.8 and 2.1 to 5.6, respectively. The average TFs of ^{210}Pb in caps and stalks were less than 1, i.e. between 0.05 to 0.35 and 0.04 to 0.83, respectively.

4. Health aspects of radioactive contamination

Radiocaesium is one of the most important anthropogenic contributors to the internal dose for humans (UNSCEAR, 1993). In Poland, there are recommendations about the activity concentration of some radionuclides in food. The tolerance limit set, especially for ^{134}Cs and ^{137}Cs in foodstuffs rarely consumed or in small

Table 3
Cap-stalk activity ratios (C/S) and transfer factor values (TF^C – cap/soil, TF^S – stalk/soil) from soil to *Xerocomus badius* (Fr.) Kühn. ex Gilb. from selected forest areas of Poland

Sample location	$\bar{x}$ ± standard deviation (range)								
	^{137}Cs			^{40}K			^{210}Pb		
	C/S	TF^C	TF^S	C/S	TF^C	TF^S	C/S	TF^C	TF^S
Augustowska Forest	1.2 ± 0.4 (1.0–1.9)	103 ± 28 (68–126)	86 ± 27 (66–121)	1.4 ± 0.7 (0.6–2.3)	3.4 ± 1.3 (1.5–4.5)	2.6 ± 0.7 (1.7–3.2)	1.9 ± 0.9 (1.2–3.3)	0.09 ± 0.02 (0.05–0.11)	0.04 ± 0.01 (0.03–0.06)
Bialowieska Forest	1.5 ± 0.2 (1.2–1.8)	34 ± 23 (11–65)	21 ± 13 (8.3–36)	1.7 ± 1.2 (0.5–3.3)	2.9 ± 1.1 (1.5–4.3)	2.2 ± 0.9 (0.9–2.9)	0.7 ± 0.4 (0.3–1.0)	0.05 ± 0.02 (0.02–0.09)	0.10 ± 0.08 (0.03–0.26)
Borecka Forest	1.1 ± 0.5 (0.2–1.6)	67 ± 71 (9.0–195)	55 ± 38 (9.9–118)	1.4 ± 0.6 (0.8–2.6)	3.1 ± 0.4 (2.5–3.8)	2.5 ± 0.8 (1.4–3.4)	0.2 ± 0.1 (0.1–0.4)	0.20 ± 0.1 (0.10–0.40)	0.83 ± 0.4 (0.34–2.4)
Adjacent area of Morag	1.8 ± 0.4 (1.2–2.2)	131 ± 94 (34–254)	89 ± 72 (16.5–170)	2.5 ± 0.6 (1.9–3.1)	4.8 ± 2.3 (2.4–7.1)	2.1 ± 1.3 (0.8–3.8)	0.5 ± 0.2 (0.2–0.7)	0.30 ± 0.2 (0.07–0.40)	0.5 ± 0.1 (0.3–0.7)
Wdzydzki Landscape Park	2.3 ± 0.6 (1.5–3.1)	137 ± 26 (120–175)	63 ± 21 (41–84)	0.8 ± 0.3 (0.5–1.2)	4.4 ± 1.1 (3.7–6.0)	5.6 ± 1.3 (4.0–6.9)	2.9 ± 2.9 (0.9–6.2)	0.35 ± 0.1 (0.31–0.43)	0.2 ± 0.1 (0.07–0.40)
Adjacent area of Kolobrzeg	1.1 ± .3 (0.8–1.4)	75 ± 51 (32–148)	63 ± 28 (36–102)	0.6 ± 0.1 (0.4–0.8)	3.1 ± 1.1 (2.2–4.2)	5.2 ± 1.0 (4.4–6.6)	0.6 ± 0.7 (0.2–1.8)	0.15 ± 0.1 (0.04–0.38)	0.6 ± 0.7 (0.03–1.59)

amounts is 1250 Bq kg^{-1} (Dziennik Ustaw No. 9, poz. 72, 2001).

Based on the average activity concentrations of 5800 Bq kg^{-1} d.w. for ^{137}Cs and 13 Bq kg^{-1} for ^{210}Pb in equilibrium with ^{210}Po, calculated including all data and an average annual consumption rate of 10 kg of fresh mushrooms, the incurred effective equivalent radiation dose would be equal to 0.08 and 0.01 mSv for ^{137}Cs and ^{210}Pb, respectively. These values are negligible compared to the permissible dose limit of 1 mSv $year^{-1}$ for the relevant critical groups of the public (IAEA, 1996). Assessments of doses were only carried out for the hypothetical population group because there is no information on their total consumption in Poland. However, consumption of wild-growing mushrooms in some regions of Poland is higher because many species are very popular and often used for garnishing dishes to improve their aroma and taste. They are collected in the forested areas, especially by villagers who are likely to eat more mushrooms than an average consume. Also, some species of wild growing mushrooms are available on the market during all seasons as dried, frozen or marinated products. According to Gaso et al. (2000), mushrooms present higher ^{137}Cs levels than those reported for other agricultural products and were shown to contribute 37% to the ^{137}Cs annual intake in Mexico. Since Cs-137 activity tends to increase along the trophic chain it is necessary to control the activity concentration of radionuclides in mushrooms, game meat and other forest fruits to evaluate radiation exposure of some groups of people and to assess its impact on human health.

Regional preferences in consumption of some food products play an important role in radionuclides intake by humans. For example, consumption of selected mushrooms species may be a major source of the intake of radiocaesium. But, most of the radiocaesium can be removed from edible mushrooms by cooking and blanching (Skibniewska & Smoczyński, 1999), thereby decreasing radiocaesium intake by humans.

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