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Mercury Content and Its Bioconcentration Factors in Wild Mushrooms at Łukta and Morąg, Northeastern Poland

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Total concentrations of mercury were determined using cold-vapor atomic absorption spectroscopy (CV-AAS) in the fruiting bodies of 16 species of wild mushrooms and underlying soil (0−10 cm) substrates collected in the areas of the Communes of Morąg and Łukta in the county of Ostróda in northeastern Poland in 1997−1998. A total of 174 composite samples of caps, 174 stalks, 80 whole fruiting bodies (collectively 1254 specimens), and 252 soils were examined. Among several species of mushrooms analyzed, the greatest concentrations were between 1300 and 71000 ng·g^{-1} of dry matter. These levels were found in the caps of Sweating mushroom (*Clitocybe rivulosa*), King Bolete (*Boletus edulis*), and Common Puffball (*Lycoperdon perlatum*) and also were characterized by the highest bioconcentration factors (BCF) for Hg, which ranged between 160 ± 82 and 110 ± 34. The cap to stalk quotient for mercury concentrations was ∼2 for most of the species except Poison Pax (*Paxilus involutus*), which had a greater concentration in caps than in stalks and a quotient of 4.4 ± 7.2. Hg concentrations in the underlying soil substrates (0−10 cm layer) ranged between 21 ± 21 and 390 ± 130 ng·g^{-1} of dry matter. The results showed that the consumption of mushrooms, considered to be the sole dietary source of mercury at the highest or mean element concentrations found, is not hazardous at daily ingestion rates of less than 70 and 210 g of fresh product, which would result in a hazard index value of less than unity.

KEYWORDS: Mercury; fungi; mushrooms; food; heavy metals

INTRODUCTION

Mercury is a toxic element to humans and wildlife, and alkylmercury compounds such as methylmercury and ethylmercury are extremely toxic. Vapors of both elemental mercury and methylmercury are spread through the atmosphere on a global scale (*1*). Thus, mercury is present in environments far from the original release sources. Fungi, including the higher fungi (higher mushrooms and macromycetes) are important organisms involved in the cycling of mercury in the terrestrial environment, especially in forest ecosystems. The presence of elevated concentrations of mercury in the flesh of edible wild mushrooms can pose threats to human health (*2*). Among the large number of higher fungi, some species have the ability to accumulate metallic elements including mercury at relatively great concentrations (*3*−*15*). The process of heavy metals uptake by mycelium from the colonized substrate and further translocation to fruiting bodies depends on many factors including the species, ecology, and other environmental features such as soil or other substrate composition and texture, degree of soil pollution, pH, humidity, and climatic conditions (*8*). In the case of mercury, it is known that some species of mushrooms (genera *Calocybe*, *Agaricus*, *Lepista*, *Macrolepiota*, *Boletus*, and *Lycoperdon*) can accumulate great concentrations even when grown in less polluted areas (*7*, *8*, *16*). Extraordinarily great concentrations of mercury have been found in the mushroom flesh collected from mercury-contaminated sites. For example, mercury concentrations in the fruiting bodies of fungi collected near a mercury plant in Idrija and Bela in the former Yugoslavia were up to 45000 ng·g^{-1} of dry matter (*17*), whereas in King Bolete mushroom (*Boletus edulis*) from an area adjacent to a mercury plant in Krompachy (Czech Republic) Hg concentrations were up to 32000 ng·g^{-1} of dry matter (*3*).

In many European countries edible mushroom picking has a long tradition and is very popular. Although the consumption rates of wild-growing mushrooms are relatively small, some people in rural areas consume large amounts (*2*), which can constitute, at least periodically, a substantial contribution to dietary intake and can be an important source of toxic metallic elements in the total diet.

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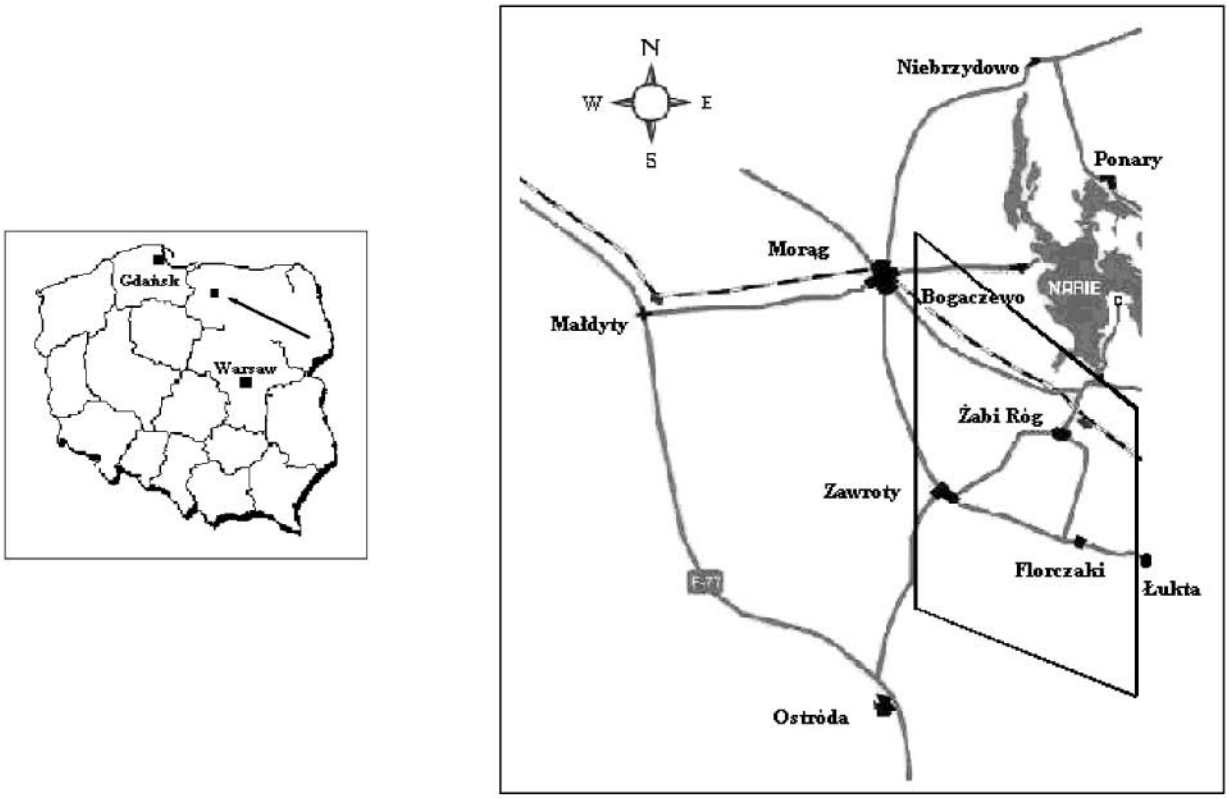

Figure 1. Area of mushroom collection at the Communes of Łukta and Morąg.

The Communes of Morąg and Łukta are largely rural and forested regions without industrial and urban centers, and the region is considered to be unpolluted with heavy metals or other chemicals. Therefore, it is considered to be a good site for harvesting wild mushrooms and other forest food resources.

This study is a part of a comprehensive investigation to elucidate the degree of mercury contamination in higher mushrooms and the underlying substrate to understand contamination status, spatial similarities, bioindication potential, and possible human intake rates and risks to local consumers in Poland.

MATERIALS AND METHODS

Fruiting bodies of 16 species of edible and inedible mushrooms as well as underlying (0−10 cm) soil samples were collected from unpolluted areas of the Communes of Morąg and Łukta in the county of Ostróda in Warminsko-Mazurskie Voivodeship in northeastern Poland (**Figure 1**) during the summer and autumn of 1997−1998. The mushroom species examined belong to the families Amanitaceae, Boletaceae, Cantharellaceae, Cortinariaceae, Humariaceae, Lycoperdaceae, Paxillaceae, Phallaceae, Russulaceae, Sclerodermataceae, and Tricholomataceae. Individual specimens of mushroom species were collected to cover a large area of land—up to several square kilometers—and to avoid excessive local sampling. Usually 15−16 pooled samples were prepared per species, and 2−10 specimens were used to make one pooled sample (**Table 1**). The total mercury concentration was determined separately in 174 composite samples of caps, 174 stalks, 80 fruiting bodies (altogether 1254 specimens), and 252 samples of underlying soil substrate (0−10 cm layer).

Fresh mushrooms were cleaned using a plastic knife, air-dried for several days and dried by heat at 40 °C for 48 h, and pulverized in an agate mortar. Subsamples (0.2−0.3 g) of dried and powdered composite samples were wet digested in 6 mL of nitric acid (Suprapoor, Merck) in closed PTFE vessels and placed in a microwave oven (Automatic Digestion System, MLS 1200). The digest was than diluted to 10 mL using double-distilled water. Two blank samples were analyzed daily for every set of up to 50 mushroom samples digested. The method of mercury measurement was validated during analyses, and appropriate data were published elsewhere (*7*, *14*, *18*).

Clean soil samples were air-dried at room temperature for ∼4 weeks and pulverized in an agate mortar and again dried in a 40 °C in oven for 48 h. The samples were further digested using a mixture of nitric and sulfuric acids in a glass system consisting of a round-bottom flask, partial condenser (30 cm long) (*7*), and water cooler. Final determination of total mercury content in mushrooms and soil was conducted using cold-vapor atomic absorption spectroscopy (CV-AAS) with an automated mercury monitor (Mercury monitor 3200, Thermo Separation Products). Analysis of the mercury content of certified reference materials (IAEA Baltic sediment SD-N-1/2) with the certified values of mercury of 1.51 $\mu g \cdot g^{-1}$ of dry matter and 1.41−1.56 confidence interval at the 0.05 significance level gives the value of 1.48 ± 0.03 $\mu g \cdot g^{-1}$ of dry matter for four determinations.

Relationships between the mercury content of the underlying soil substrate versus the mercury content of dried caps, stalks, and/or whole fruiting bodies as well as bioconcentration factor (BCF) values of mercury against substrate concentration were examined by linear regression analysis.

RESULTS AND DISCUSSION

Concentrations of mercury in fungi varied depending on the species examined (**Table 1**). The greatest concentrations of mercury were found in the flesh of Sweating mushroom (*C. rivulosa*) of up to 5200 ± 1500 $ng \cdot g^{-1}$ of dry matter in the caps and 1900 ± 900 $ng \cdot g^{-1}$ of dry matter in the stalks, followed by King Bolete (*B. edulis*) with 3000 ± 1600 and 1800 ± 900 $ng \cdot g^{-1}$ and Common Puffball (*L. perlatum*) with 2800 ± 500 $ng \cdot g^{-1}$. The Common Earth Ball (*S. citrinum*) had the lowest level of mercury with 9.3 ± 3.0 $ng \cdot g^{-1}$ of dry matter.

The caps usually contained a higher concentration of mercury than the stalks, and cap to stalk concentration quotients for mercury ranged from 1.5 ± 0.7 for Red-capped Scaber Stalk (*L. rufum*) to 4.4 ± 7.2 for Poison Pax (*P. involutus*).

Because mercury is one of the most toxic elements, regulations for mercury levels in many kinds of foodstuffs are enforced

Table 1. Mercury Concentrations in Mushrooms and Soil Substrate (Nanograms per Gram of Dry Weight), Hg Bioconcentration Factors (BCF) in Caps and Stalks, and Cap to Stalk (C/S) Hg Concentration Quotients

mushroom species	no. of samples	cap Hg	BCF	stalk Hg	BCF	C/S	soil Hg
Bay Bolete	16 (44)[a]	140 ± 84; 140[b]	7.3 ± 4.8; 3.2	72 ± 42; 64	3.3 ± 2.6; 2.2	1.9 ± 0.6; 1.8	25 ± 15; 24
Xerocomus badius (Fr.) Kühn. ex Gilb.		(77–310)	(1.4–16)	(12–140)	(0.55–9.3)	(1.2–3.5)	(12–67)
King Bolete	16 (24)	3000 ± 1600; 3300	140 ± 110; 190	1800 ± 900; 1300	79 ± 57; 90	1.9 ± 0.8; 1.9	21 ± 21; 18
Boletus edulis Bull.: Fr.		(1800–7100)	(57–430)	(920–3700)	(21–210)	(1.2–3.7)	(7.0–100)
Common Scaber Stalk	16 (26)	700 ± 270; 460	8.7 ± 5.4; 5.7	350 ± 140; 250	4.3 ± 2.5; 3.3	2.0 ± 0.5; 1.7	90 ± 83; 89
Leccinum scabrum (Bull.: Fr.)		(140–1400)	(1.6–22)	(98–660)	(0.78–9.6)	(1.0–2.8)	(24–360)
Red-capped Scaber Stalk	16 (28)	600 ± 590; 1100	28 ± 30; 45	450 ± 390; 650	20 ± 26; 22	3.0 ± 5.4; 1.3	25 ± 12; 24
Leccinum rufum (Schaeff.) Kreisel		(290–2400)	(10–100)	(62–1800)	(3.6–95)	(1.1–23)	(11–55)
Poison Pax	16 (34)	270 ± 260; 98	10 ± 10; 5	90 ± 110; 43	3.7 ± 5.9; 1.7	4.4 ± 7.2; 2.8	52 ± 41; 32
Paxillus involutus (Batsch.: Fr.) Fr.		(13–710)	(0.3–27)	(10–420)	(0.16–7.4)	(1.3–29)	(8.0–380)
Common Chantarelle	16 (162)[c]	27 ± 14; 28	0.33 ± 0.88; 0.7				54 ± 94; 34
Cantharellus cibarius Fr.		(10–54)	(0.072–3.6)				(10–390)
Soap-scented Knight-cap	15 (87)	34 ± 20; 40	0.81 ± 0.55; 1.1	15 ± 8; 16	0.30 ± 0.23; 0.45	2.3 ± 1.0; 2.6	41 ± 6; 36
Tricholoma saponaceum		(9.0–66)	(0.26–1.9)	(5.0–35)	(0.14–1.1)	(1.1–4.3)	(28–49)
Gray Knight-cap	15 (92)	25 ± 34; 32	0.64 ± 0.81; 0.77	14 ± 17; 17	0.39 ± 0.33; 0.47	2.2 ± 0.7; 2.0	42 ± 16; 40
Tricholoma terreum		(23–150)	(0.44–3.3)	(9.0–76)	(0.15–1.2)	(1.1–3.2)	(22–79)
Fly Agaric	16 (48)	870 ± 220; 700	15 ± 9; 17	390 ± 190; 350	5.4 ± 4.9; 8.3	2.5 ± 0.7; 1.9	150 ± 70; 39
Amanita muscaria (L.) Pers.		(290–1000)	(3.5–34)	(110–750)	(1.9–19)	(1.1–3.8)	(19–280)
Red-hat Milk Cap	16 (75)	45 ± 22; 29	0.51 ± 0.22; 0.24	17 ± 13; 20	0.19 ± 0.10; 0.16	2.7 ± 1.3; 1.7	110 ± 70; 39
Lactarius rufus		(14–83)	(0.088–0.85)	(5.0–48)	(0.015–0.33)	(1.0–5.8)	(19–280)
Fairy-ring mushroom	16 (124)	730 ± 390; 580	19 ± 10; 10	480 ± 210; 360	13 ± 6; 8	1.5 ± 0.7; 1.6	39 ± 48; 48
Marasimus oreades		(230–1700)	(2.4–40)	(130–830)	(0.82–20)	(1.0–3.5)	(20–170)
Sweating mushroom	16 (54)	5200 ± 1500; 4200	160 ± 82; 160	1900 ± 900; 2300	56 ± 45; 81	2.8 ± 0.7; 1.9	37 ± 14; 30
Clitocybe rivulosa		(2300–7000)	(33–310)	(820–4100)	(22–170)	(1.3–4.0)	(11–70)
Common Puffball	16 (138)[c]	2800 ± 490; 2200	89 ± 48; 78				54 ± 120; 26
Lycoperdon perlatum		(1300–3100)	(6.6–210)				(12–470)
Common Earth Ball	16 (112)[c]	9.3 ± 3.0; 13	0.026 ± 0.076; 0.058				380 ± 130; 220
Scleroderma citrinum		(9.0–21)	(0.019–0.24)				(55–480)
Orange Peel fungus	16 (168)[c]	47 ± 49; 33	1.5 ± 2.6; 1.3				39 ± 13; 20
Aleuria aurantia		(4.0–190)	(0.083–7.3)				(8.0–55)
Shameless Stinkhorn	16 (38)[c]	350 ± 330; 590	1.8 ± 3.5; 3.4				200 ± 60; 190
Phallus impudicus		(320–1400)	(1.7–12)				(43–250)

[a] Number of pooled samples and total number of fruiting bodies (in parentheses). [b] Median value. [c] A whole fruiting body.

in Poland. The tolerance limit of mercury in fresh edible mushrooms is 50 ng·g^{-1}, whereas it is 500 ng·g^{-1} in dried mushrooms (*20*). On this basis, mercury concentrations in most samples of dried edible mushroom species examined in this study were above the legal tolerance limit mentioned. Nevertheless, when concentrations were considered on a fresh weight basis (moisture content of 90%, on average), all species, except Bay Bolete (*X. badius*), Common Chantarelle (*C. cibarius*), and Gray Knight-cap (*T. terrerum*), had concentrations in excess of the threshold limits (**Table 1**).

Wild-grown mushrooms are traditional food items in Polish cuisine and in other European culinary cultures and are still popular at the present time. In the Czech Republic, 72% of families pick mushrooms at a mean annual rate of 7 kg per household, whereas for some individuals consumption rates exceed 10 kg per annum (*2*). There are no statistical data available on the collection and/or consumption rates of wild or cultivated mushrooms in Poland, and their picking is a very popular activity among Poles. The rates of mushroom picking and estimated consumption are similar to those in the Czech Republic. The consumption of fresh mushrooms by villagers can be periodically high, especially during the summer and autumn. Additionally, dried specimens of several mushroom species, for use in soups, sauces, and bouillon and as ground mushroom species, are commercially widely available at all seasons.

Noncarcinogenic health effects may be estimated using a mercury reference dose value (RfD) of 0.0003 mg·kg^{-1}·day^{-1} (*21*, *22*). The RfD is an estimate for a single daily intake rate that may be without risk if ingested over a lifetime. The estimated dose (D) can be calculated as $D = C \times I/W \times 1000$,

Table 2. Hazard Index for Ingestion of Mushrooms Containing Mercury (Łukta and Morąg Sites)

mushroom consumption rate (g·day^{-1})	hazard index (H): based on greatest Hg concn (0.30 μg·g^{-1} of wet wt)	hazard index (H): based on mean Hg concn (0.10 μg·g^{-1} of wet wt)
6.4	0.09	0.03
28	0.40	0.13

Table 3. Mercury Concentrations (Nanograms per Gram of Dry Matter) in Underlying Soil (0–10 cm) of Wild Mushrooms

soil substrate type	no. of samples	range	mean ± SD
all samples	252	3.0–480	250 ± 90
sand with light clay and light humus	98	3.0–470	360 ± 60
sand with light clay and heavy humus	44	12–360	120 ± 78
sand with heavy clay	32	8.0–390	23 ± 68
sand	48	11–120	39 ± 25
organic matter	30	47–480	180 ± 94

where C is the concentration of mercury in mushroom (μg·g^{-1} of wet wt), I is the ingestion rate of mushroom (g·day^{-1}), and W is average body weight (70 kg). The hazard index (H) for a substance is the ratio of dose (D) to the upper daily level substance intake rate over a lifetime and is estimated to be without toxic effects (i.e., RfD). If the H value is <1, toxic effects will not occur. The H value can be calculated as a function of ingestion rate and concentration of mercury ion in mushrooms. The ingestion rates were chosen to represent

Table 4. Relationship between the Mercury Content of Mushrooms (Caps, Stalks, or Whole Fruiting Body) and the Underlying Soil (0–10 cm) as well as between the BCF and the Size of the Entire Fruiting Bodies[a]

	specific ratios			
mushroom species	Hg_C/Hg_{US}	Hg_{St}/Hg_{US}	BCF_C/cap_D	$BCF_{St}/stalk_H$
King Bolete *B. edulis*	0.39	0.19	–0.52*	–0.47
Bay Bolete *X. badius*	0.14	0.16	–0.28	–0.52*
Common Scaber Stalk *L. scabrum*	0.19	0.24	–0.51*	–0.59*
Red-capped Scaber Stalk *L. rufum*	0.14	–0.084	–0.53*	–0.50*
Poison Pax *P. involutus*	–0.15	0.31	–0.39	–0.31
Common Chantarelle *C. cibarius*	0.028		–0.36	
Soap-scented Knight-cap *T. saponaceum*	0.17	0.12	–0.19	–0.20
Gray Knight-cap *T. terreum*	0.20	0.26	–0.34	–0.25
Fly Agaric *A. muscaria*	0.55*	0.50*	–0.64**	0.53*
Red-hot Milk Cap *L. rufus*	0.0031	0.049	–0.46	–0.51*
Fairy-ring mushroom *M. oreades*	0.01	–0.071	–0.55	–0.65*
Sweating mushroom *C. rivulosa*	–0.28	–0.18	–0.85**	–0.76**
Common Puffball *L. perlatum*	0.54*		–0.28	
Shameless Stinkhorn *Ph. impudicus*	–0.13		–0.87**	
Common Earth Ball *S. citrinum*	–0.29		–0.82**	
Orange Peel fungus *A. aurantia*	–0.039		–0.36	

[a] C, cap; St, stalk; US, underlying substrate; BCF, bioconcentration factor; D, diameter in cm of the cap; H, height of the stalk; *, $p < 0.05$; **, $p < 0.01$.

average consumption rates per household member (6.4 g·day^{-1}) in the region of the Communes of Łukta and Morąg (assuming 7 kg annually per three-member family) and the highest consumption rate per individual (28 g·day^{-1}, assuming 10 kg annually). The *H* values were calculated for the ingestion of mushrooms containing the greatest concentration of 300 ng·g^{-1} of wet wt for King Bolete with a mean value of 100 ng·g^{-1} of wet wt for the caps of all edible species (**Table 2**). The results also showed that mushroom consumption at the highest or mean mercury concentrations is not hazardous at ingestion rates of less than 70 and 210 g of fresh product day^{-1}, respectively, which would result in a hazard index value of less than unity.

Soil mercury concentrations ranged between 21 ± 21 ng·g^{-1} of dry matter for King Bolete (*B. edulis*) and 380 ± 130 ng·g^{-1} for Common Earth Ball (*S. citrinum*) as shown in **Table 1**. The total mean mercury concentration for soil was 250 ± 90 ng·g^{-1} of dry matter, whereas the greatest values were found in soil samples with greater portions of humus than in sandy soils (**Table 3**).

Quotients for mushroom to soil mercury concentration were calculated to clarify the bioconcentration potential of this element (preferable uptake of mercury from soil) by the species examined. The bioconcentration factors (BCFs) of total mercury were very high in Sweating mushroom (*C. rivulosa*), King Bolete (*B. edulis*), and Common Puffball (*L. perlatum*) as shown in **Table 1**. The range of BCF values for two of these mushroom species were 160 ± 82 and 140 ± 110 for the caps and 79 ± 57 and 56 ± 45 for the stalks, respectively, whereas the BCF was 110 ± 34 for whole fruiting bodies of Common Puffball (*L. perlatum*) as also recorded in **Table 1**. Some of the mushroom species examined contained low Hg BCF values of <1, which can be considered to be mercury excluders. The excluders are such species as Common Earth Ball (*S. citrinum*), with an average Hg^{2+} BCF value as low as 0.026 ± 0.076 (median value = 0.058), Common Chantarelle (*C. cibarius*), with 0.33 ± 0.88, Red-hat Milk Cap (*L. rufus*), with 0.51 ± 0.22, Gray Knight-cap (*T. terreum*), with 0.64 ± 0.81, and Soap-scented Knight-cap (*T. saponaceum*), with 0.81 ± 0.55 for the caps.

Some of these mushroom species grew in soil with somewhat higher mercury concentrations when compared to other species; for example, soils underlying Common Earth Ball (*S. citrinum*) and Red-hat Milk Cap (*L. rufus*) had mercury concentrations of 110 ± 70 and 380 ± 130 ng·g^{-1} of dry matter. However, these two species were unable to accumulate considerable amounts of this element (**Table 1**). On the contrary, species such as Fairy-ring mushroom (*M. oreades*), King Bolete (*B. edulis*), and Common Puffball (*L. perlatum*) accumulated elevated concentrations of this element, which can probably be explained by their species-specific potential to bioconcentrate mercury.

Regression analysis of the relationship between mercury concentrations in mushrooms and the corresponding soil substrate revealed that the levels in the caps and stalks of Fly Agaric (*A. muscaria*) and in the whole fruiting bodies of Common Puffball (*L. perlatum*) increased ($p < 0.05$) with increasing soil mercury concentrations, whereas no trend could be observed for other species analyzed (**Table 2**). Interestingly, Hg BCF values for many species decreased ($0.01 < p < 0.05$) with an increase in the diameter of the cap or in the height of the stalk (**Table 2**).

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