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Mercury Bio-Concentration Potential of Larch Bolete, *Suillus grevillei*, Mushroom

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Abstract Larch Bolete can be classified as a mushroom species accumulating Hg in the fruiting bodies. Our results did indicate diversity of Hg concentrations ($p < 0.05$), depending on a site of collection of Larch Bolete fruiting bodies as well as the lack of any statistically significant differences in soil mercury contamination among the examined sites. Values of 1.0 μg Hg/g dw for pool of caps and 2.0 μg/g dw for a single cap of Larch Bolete are suggested as threshold concentrations implying Hg baseline level, while greater value could imply contamination due to the site pollution.

Keywords Bioconcentration · *Suillus grevillei* · Metals · Mushroom · Soil

Mercury remains an environmentally problematic element. Long-range atmospheric transport of elemental mercury vapours caused that it is difficult to consider wildlife forest areas as unaffected by this metal emissions. Forest soil, which is rich in organic matter substratum for fungus mycelia and easily absorbs mercury from both air deposition and weathering of parent bedrock, can be described as "Hg reservoir" (Suchara and Sucharová 2002).

Total mercury content of mushrooms is species dependent and varies amongst their ecological groups (saprophytic, symbiotic, etc.), which hinders the interpretation of data for different species and areas (Alonso et al. 2000). Owing to some ability to bio-concentrate mercury, certain mushroom species can find application in monitoring the pollution of forest areas with this element. Larch Bolete mushroom is relatively common edible species that springs up in early autumn near or under its host trees of Larch (*Larix decidua*). This mushroom is easy to recognize and popular among mushroom fanciers.

Aim of this study was to investigate the distribution of total Hg concentration in Larch Bolete fruiting bodies relevant to humans.

Materials and Methods

Fruiting bodies (carpophores) of Larch Bolete *Suillus grevillei* (Klotzch: Fr) Singer and underlying (0–10 cm) soil substrate were collected from nine, spatially distant sites in Northern part of Poland within 2000–2006. Carpophores were collected from the Sępopolska Lowland, Elbląska Upland, Kaszuby Lake District, Tucholskie Forest, Polanowska Upland, Staropruskie Coast, Vistula sandbar, Kaszubskie Coast and Hel Peninsula in Poland. Mycelia of *Suillus grevillei* different genets can be distributed over the wide range of depths in soil substrate (Zhou et al. 2001) Sampling at 0–10 cm depth horizon aimed to recognize spatial variations in Hg soil substrate content, assess Hg accumulation potential of this species and to examine relationships between mercury contamination of soil substrate and Larch Bolete fruiting bodies.

For each site, from ten to fifteen samples of specimens were collected. One sample consisted of two or three carpophores. After cleanup from the plant material and particles of soil with a plastic knife, an initial air-drying in room temperature in well-ventilated place for 2–3 days, fruiting bodies were further oven dried at 80°C for 48–96 h, and next crushed and ground in agate mortar to fine powder.

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Mercury content of soil from particular sites was also determined in pooled samples. Each of them was made of soil substratum from two or three spatially nearest fruiting bodies sampling points, using soil amount equivalent by weight (5 g). The samples, after removal of the visible organisms, small stones, sticks and leaves were air dried in room temperature for approximately 4 weeks and then sieved through a pore size of 2 mm.

Total mercury content of fruiting bodies and soil substrate underneath them has been determined by cold-vapour atomic absorption spectroscopy (CV–AAS) after thermal decomposition of sample matrix and further amalgamation and desorption of element from gold wool (Mercury analyzer type MA-2000, Nippon Instruments Corporation, Takatsuki, Japan). Analytical control and quality assurance (AC/QA) have been performed through analysis of certified reference material CS-M-1 (dried fruiting bodies of mushroom Cow Bolete *Suillus bovinus*), produced by the Institute of Nuclear Chemistry and Technology in Warsaw, Poland. Declared content of total mercury in CS-M-1 reference material was 0.174 ± 0.018, while our measurements showed 0.171 ± 0.008 μg/g dw (n = 3). In addition, with every set of 10 mushrooms or soil samples, one blank sample was examined. For mushroom and soil substrate, the method limit of detection (LOD) was 0.005 μg Hg/g dw, and the quantification limit (LOQ) was 0.003 μg Hg/g dw.

Mercury concentration in fruiting bodies has no Gaussian distribution. Data transformation, which aimed to obtain their log-normal distribution, was unsuccessful. Moreover, data variances were heterogeneous (Barlett test). Consequently, statistical analyses were performed with nonparametric tests. The same applies with respect to soil Hg concentration. Wilcoxon matched pairs test was used to describe a statistical significance of the differences between Hg content of caps and stipes, and Spearman's Rank Correlation was used to test the relationship between them. Analysis of variance (Kruskal–Wallis test) was performed to indicate possible and site dependent diversity in Hg concentrations. Further examination of differences in Hg concentrations between the caps (also stipes and soil) from certain localizations was made using Dunn post test.

Results and Discussion

The arithmetic means of the Larch Bolete caps' mercury concentration varied, depending on a site, between 0.26 ± 0.08 and 0.50 ± 0.10 (Fig. 1), and for stipes were between 0.09 ± 0.03 and 0.16 ± 0.07 μg/g dw. Maximum mercury concentration noted in a single cap and stipe was up to 0.84 and 0.28 μg/g dw, respectively (Table 1). Depending on the site, the mushroom's caps were from 2.4 ± 0.7 to 4.0 ± 0.5-fold more affected with mercury when compared to stipes ($p < 0.05$). An overall mean of cap to stipe Hg concentration quotient for all 121 specimens of Larch Bolete examined was 3.1 ± 1.1. Relationships between mercury concentrations of caps and stipes were positive ($r = 0.67$) and statistically significant ($p < 0.05$).

In the case of mercury and Larch Bolete mushroom, analysis of variance indicated some diversity of concentrations ($p < 0.05$) depending on a site (place) of their origin. Further statistical evaluation indicated that mercury content of caps varied ($p < 0.05$) between certain sites

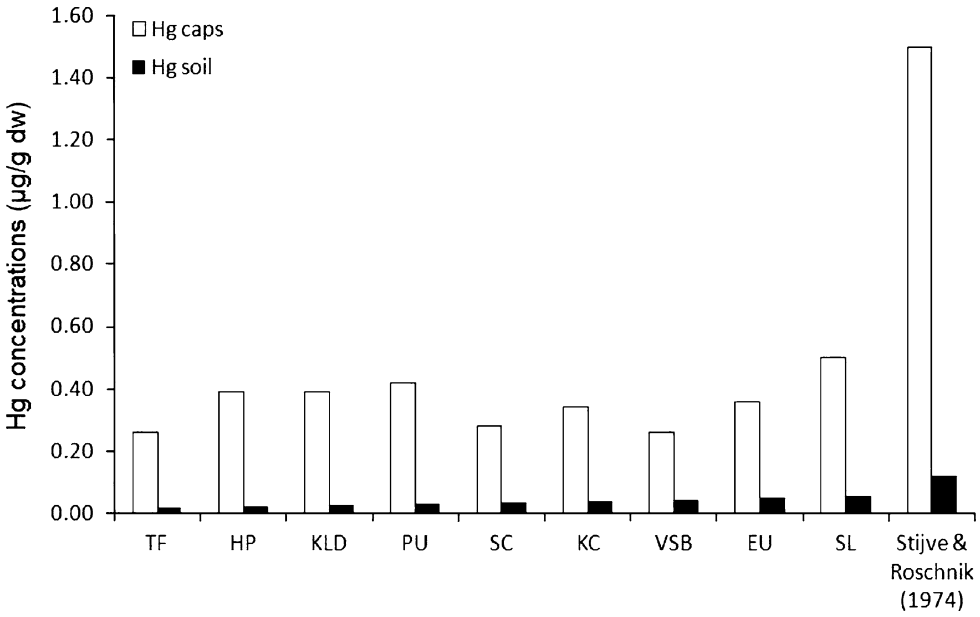

Fig. 1 Mean Hg concentration of soil substrate and caps of Larch Bolete; the sampling sites are displayed in order from small to greater soil Hg concentration. Abbreviations: *TF* Tucholskie Forest, *HP* Hel Peninsula, *KLD* Kaszuby Lake District, *PU* Polanowska Upland, *SC* Staropruskie Coast, *KC* Kaszubskie Coast, *VSB* Vistula sand bar, *EU* Elbląska Upland, *SL* Sępopolska Lowland, Stijve and Roschnik (1974) – literature data

Table 1 Total mercury concentrations in caps and stipes of Larch Bolete and underlying soil (μg/g dw), BCF and cap to stipe Hg content ration quotients ($Q_{C/S}$)

No.	Sample location and year	Cap	Stipe	Soil	BCF_C	BCF_S	$Q_{C/S}$
1	Hel Peninsula 2006 (15;5)[a]	0.39 ± 0.19 (0.40)[b] 0.12–0.68	0.16 ± 0.07 (0.13) 0.075–0.28	0.019 ± 0.003 (0.020) 0.014–0.021	21 ± 10 (21) 6.6–36	8.5 ± 3.7 (7.2) 4.0–15	2.4 ± 0.7 (2.2) 1.5–3.9
2	Kaszubskie Coast 2004 (15;5)	0.34 ± 0.16 (0.32) 0.13–0.60	0.12 ± 0.03 (0.11) 0.077–0.19	0.035 ± 0.004 (0.035) 0.030–0.041	9.6 ± 4.5 (11) 3.8–17	3.3 ± 1.0 (3.2) 2.2–5.3	2.8 ± 0.7 (2.9) 1.7–3.7
3	Vistula sand bar 2004 (11;5)	0.26 ± 0.08 (0.25) 0.15–0.39	0.10 ± 0.03 (0.094) 0.064–0.14	0.041 ± 0.010 (0.044) 0.026–0.048	6.5 ± 2.0 (6.1) 3.7–9.6	2.5 ± 0.6 (2.3) 1.6–3.5	2.6 ± 0.2 (2.7) 2.3–2.8
4	Polanowska Upland 2003 (15;5)	0.42 ± 0.04 (0.41) 0.36–0.49	0.11 ± 0.02 (0.10) 0.083–0.13	0.027 ± 0.006 (0.025) 0.021–0.037	16 ± 1 (16) 14–18	4.0 ± 0.6 (3.9) 3.1–5.1	4.0 ± 0.5 3.8 3.3–5.0
5	Staropruskie Coast, 2003 (10;5)	0.28 ± 0.17 (0.28) 0.10–0.54	0.12 ± 0.06 (0.13) 0.036–0.24	0.033 ± 0.012 (0.031) 0.023–0.051	8.5 ± 5.2 (8.2) 2.9–16	3.6 ± 1.9 (3.9) 1.1–7.3	3.1 ± 3.3 (2.3) 0.63–12
6	Tucholskie Forest 2002 (15;5)	0.26 ± 0.08 (0.25) 0.14–0.42	0.089 ± 0.026 (0.081) 0.051–0.15	0.016 ± 0.012 (0.011) 0.0053–0.036	16 ± 5 (16) 8.7–27	5.6 ± 1.7 (5.1) 3.2–9.2	3.1 ± 1.4 (3.1) 1.4–6.3
7	Elbląska Upland 2001 (10;5)	0.36 ± 0.17 (0.34) 0.17–0.67	0.11 ± 0.04 (0.099) 0.069–0.19	0.047 ± 0.035 (0.027) 0.020–0.11	7.8 ± 3.7 (7.4) 3.6–14	2.3 ± 0.8 (2.1) 1.5–4.2	3.7 ± 2.2 (2.7) 1.7–7.9
8	Kaszuby Lake District 2001 (15;5)	0.39 ± 0.17 (0.37) 0.17–0.69	0.11 ± 0.05 (0.10) 0.059–0.20	0.024 ± 0.003 (0.023) 0.022–0.027	16 ± 7 (16) 7.3–29	4.8 ± 1.9 (4.1) 2.5–8.2	3.4 ± 0.7 (3.4) 2.2–4.9
9	Sępopolska Lowland 2000 (15;5)	0.50 ± 0.10 (0.46) 0.38–0.84	0.15 ± 0.03 (0.14) 0.11–0.25	0.053 ± 0.024 (0.055) 0.029–0.085	9.3 ± 2.3 (8.5) 7.1–16	2.8 ± 0.7 (2.6) 2.1–4.7	3.4 ± 0.5 (3.4) 2.6–4.6

Statistically significant differences between caps: 3–9, 5–9, 6–9; statistically significant differences between stipes: 1–6, 6–9; for example: 3–9 difference between caps of fruiting bodies from Vistula sand bar and Sępopolska Lowland

[a] Number of fruiting bodies; number of pooled soil samples

[b] Median values

(Table 1). Differences noted for caps at certain sites could not always be confirmed for stipes ($p < 0.05$).

In some earlier surveys of Larch Bolete from the stands in Poland, total mercury content of caps varied between 0.22 ± 0.06 and 0.60 ± 0.10 μg/g dw, and that of stipes between 0.10 ± 0.05 and 0.26 ± 0.10 μg/g dw (Falandysz et al. 2002a, b, c, 2004). Two sets of whole fruiting bodies of Larch Bolete collected from the contaminated Middle Spiš region of Slovakia contained mercury at 1.1 ± 0.5 and 4.8 μg/g dw (Kalač et al. 1991; Zimmermannová et al. 2001), what is elevated when compared to baseline concentrations observed in this study (Table 1).

None of the surveyed sites was recognized as being under the influence of any local mercury pollution source. Total mercury content of forest soil substrate for Larch Bolete depending on the site, revealed low contamination and mean concentrations varied between 0.016 ± 0.012 and 0.053 ± 0.024; total range 0.0053–0.11 μg/g dw (Table 1). A multiple statistical comparisons indicated a lack of any significant differences in soil mercury contamination among the sites surveyed ($p < 0.05$). Soil mercury data gained in this study imply baseline concentrations there. Range of the values reported for top (0–10 cm) horizon of forest soils in a broader study conducted across Poland revealed values from 0.04 to 0.25 μg/g dw (Falandysz et al. 1996).

Based on calculated values of bioconcentration factor (BCF; called also as transfer factor, TF or enrichment factor, EF; and which is a quotient between element in mushroom to soil concentration on dry weight basis), Larch Bolete could be classified as species accumulating (BCF > 1) but not excluding (BCF < 1) mercury. For caps, means of mercury BCF, depending on the site, varied between 6.5 ± 2.0 and 21 ± 10, and for stipes between 2.3 ± 0.8 and 8.5 ± 3.7 (Table 1). In earlier reports mercury BCF values of Larch Bolete ranged from 12 ± 12 to 140 ± 76 and from 2.8 ± 2.0 to 62 ± 49 for caps and stipes, respectively (Falandysz et al. 2002a, b, c, 2004). BCF value of mercury calculated for whole fruiting body of a single specimen of Larch Bolete from a stand in Switzerland was 11 (Stijve and Roschnik 1974).

In present study, relationship between mercury content of the fruiting bodies of Larch Bolete and magnitude of soil contamination has not been tested. This is due to the lack of statistically important differences ($p < 0.05$) of soil mercury content between the forested areas surveyed. Previous investigations indicated that fruiting bodies of Larch Bolete could potentially contain a considerably larger amounts of mercury (Kalač et al. 1991; Zimmermannová et al. 2001). Based on published reports, only that of Stijve and Roschnik (1974) provides information on mercury content of mushroom and soil substrate underneath the fruiting body. Having analysed these data, it seems that Hg content of Larch Bolete's fruiting body is influenced by its soil concentration, however, direction (linear or nonlinear) and power of the dependence remain unknown. Nevertheless, it is clear that Hg bioavailability to mushroom depends on other factors as well (Fig. 1).

Results of our study suggest that Larch Bolete mushroom could be used for Hg forest soil monitoring purpose. Mushroom sampling is easy to apply compared to effort necessary to sample structurally complex forest soil. Variability in Hg content of *Suillus grevillei* fruiting bodies from the spatially distant sites has been confirmed but it has not been found for underlying soil substrate. Maximum median and mean mercury content of caps of Larch Bolete collected from the uncontaminated sites surveyed in Poland was up to 0.60 μg/g dw, and in a single cap was up to 1.6 μg/g dw (Falandysz et al. 2002a, b, c, 2004). Based on these findings, it seems reasonable to conclude that baseline mercury concentration of a pooled collection of caps of Larch Bolete should not exceed 1.0 μg/g dw, while 2.0 μg/g in a single cap. These values could be considered as threshold value for "uncontaminated" fruiting bodies of Larch Bolete, while greater mercury content could imply mushroom contamination due to elevated Hg content of forest soil and its pick-up at contaminated areas.

Wild grown mushrooms, including Larch Bolete, are traditional and at least seasonally popular food item in various countries. In Poland over 6,000 tones of fresh wild grown forest mushrooms have been collected for commercial purposes in 2000 (Grzesiak et al. 2007), while amount picked-up by non-commercial mushroom fanciers certainly could be much greater.

To assess possible risk due to intake of mercury accumulated in fruiting bodies of Larch Bolete, a reference dose (RfD; 0.0003 mg/kg bw daily) and value of Provisionally Tolerable Weekly Intake (PTWI; 0.005 mg/kg bw) (JECFA 1978; US EPA 1987) have been applied in this study.

Cap of well-grown fruiting body of Larch Bolete is much bigger by mass than stipe (over 95% of fruitbody weight). Hence, mercury content of caps is a major concern for this species. Depending on the sampling site, a meal made with 650–1,200 g of fresh caps of Larch Bolete (from the Sępolska Lowland and the Tucholskie Forests site, respectively, Table 1) will not result in exceeding mentioned mercury reference dose. In the case of PTWI value, even eating weekly up to 11 kg of fresh caps of Larch Bolete, when picked-up at the sites surveyed, will not result in exceeding this limit, provided that no mercury from other food items are not consumed.

PTWI for methylmercury is 1.6 μg/kg bw (JECFA 2007). Unfortunately, no data on methylmercury content of Larch Bolete could be obtained in this study. Stijve and Roschnik (1974) found highly toxic methylmercury at 6.6% of the total mercury content in this mushroom species.

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