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Mercury and its bioconcentration factors in Brown Birch Scaber Stalk (*Leccinum scabrum*) from various sites in Poland

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Abstract

The total mercury contents were determined in the carpophores of Brown Birch Scaber Stalk (*Leccinum scabrum*) and topsoil (0–10 cm) collected from 12 spatially distant sites across Poland. Mercury was measured by cold-vapour atomic absorption spectroscopy (CV-AAS) after nitric acid (mushrooms) or *aqua regia* (soil) digestion of the samples. The caps, depending on the site, had total mercury concentrations from 0.38 ± 0.23 to 1.2 ± 0.4 μg/g dm (median 0.36–1.2 μg/g dm), and stalks from 0.17 ± 0.08 to 0.72 ± 0.20 μg/g dm (median 0.17–0.72 μg/g dm). Overall-mean mercury contents for 240 caps and stalks were 0.63 ± 0.38 (0.072–2.0 μg/g dm) and 0.32 ± 0.20 (0.028–1.2 μg/g dm), respectively. The total mercury content in top soil layer (0–10 cm) at 12 sites, after hot *aqua regia* extraction, averaged 0.026 ± 0.010–0.066 ± 0.018 μg/g dm. The BCF values of total mercury in caps of Brown Birch Scaber Stalk from the particular sites ranged from 14 ± 5 to 20 ± 4 (total mean was 16 ± 5, and median 18), in stalks from 6.0 ± 4.0 to 11 ± 1 (total mean was 8.3 ± 3.1, and median 8.1). In human feeding, wild mushrooms are usually only a small part of the total diet, so consumption of Brown Birch Scaber Stalk collected from the background sites in Poland, or elsewhere in Europe, as regards mercury content, could be considered safe.

Keywords: BCF; Bioindication; Fungi; Heavy metals; Mineral composition; Mushrooms; Wild food

1. Introduction

Monomethylmercury ion, due to its high stability, lipophilic properties, bioavailability, bioaccumulation and biomagnification potential and neurotoxicity, as well as relative environmental abundance, is to humans the most hazardous and relevant mercury moiety (JPHS, 2001a, 2001b). In the case of the higher fungi group (mushrooms, macromycetes), which is very numerous in species, (many of these edible for humans) can contain total mercury in the carpophore (fruitbody) at surprisingly high concentration, even, if picked from unpolluted areas (Alonso, Salgado, Garcia, & Melgar, 2000; Falandysz, Gucia, Frankowska, Kawano, & Skwarzec, 2001; Falandysz et al., 2001; Falandysz et al., 2003; Tuzen & Soylak, 2005; Cocchi, Vescovi, Petrini, & Petrini, 2006). Vegetables, fruits, berries, tubers, edible roots, grains, and generally plants and wild-grown vegetable foods, as well as game and slaughtered animals or freshwater and marine fish, in Poland, usually contain total mercury at lower or much lower levels than do many wild-grown edible mushrooms (Falandysz, 1990, 1991, 1993a, 1993b, 1994a, 1994b; Falandysz, Kotecka, & Kannan, 1994; Falandysz, Chwir, & Wyrzykowska, 2000; Varo, Lähelmä, Nuurtamo, Saari, & Koivistoinen, 1980; Weeks et al., 2006). The mushroom species, which are known as efficient mercury accumulators when grown at mercury polluted sites do contain this element at strikingly high concentration, and usually more in cap than in stalk (Fischer, Rapsomanikis, Andreae, & Baldini, 1995; Kalač, Nižnanská, Bevilaqua, & Stašková, 1996). Methylmercury forms only a small portion of the total mercury found in carpophores of higher mushrooms. Nevertheless, methylmercury can be much more efficiently

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bioconcentrated by some species than inorganic mercury compounds, and some mushroom species may even be able to methylate mercury (Fischer et al., 1995).

This study is part of a comprehensive survey to investigate metallic elements and metalloids in some edible and inedible higher mushrooms and topsoil to understand contamination status, bioindication potential and risk to the local consumers in Poland. In this paper data are reported on total mercury content of the fruiting bodies of Brown Birch Scaber Stalk (*Leccinum scabrum*) and topsoil (0–10 cm), and the mercury bioconcentration factor (BCF) by this species collected from 12 spatially distant sites across Poland. Some preliminary conference data were earlier presented (Bielawski & Falandysz, 2003).

2. Materials and methods

2.1. Samples

Carpophores of Brown Birch Scaber Stalk (*Leccinum scabrum*) (Bull.: Fr.) S.F. Gray and underlying soil substrate (top 0–10 cm layer; ~100 g) were collected from twelve spatially distant sites in Poland in 1998–2001 (Fig. 1). Individual, well grown and roughly similar in size specimens of Brown Birch Scaber Stalk from each site were collected over a relatively large area of forest to avoid excessive local sampling.

2.2. Analysis

The total mercury content was determined separately in the caps and stalks of Brown Birch Scaber Stalk and in (underlying the carpophore) soil substrate (0–10 cm layer). Fresh mushrooms, after cleanup from the plant and substrate debris with a plastic knife, were air-dried for several days and further dried in an oven at 40 °C for 48 h, and pulverized in an agate mortar. A sub-sample of powdered mushroom (~0.5 g) was wet-digested with 6 ml of concentrated nitric acid (Suprapoor®, Merck) in a closed PTFE vessel in a microwave oven (MARS 5 of CEM Corp., Matthews, NC, USA). With every set of up to 50 mushroom samples, digested daily, 5 duplicates and two blank samples were run. The digest was further diluted using deionised water.

The soil samples, after removal of plants, small stones and visible organisms, were dried at clean conditions for a few weeks at room temperature and further in an electric oven at 40 °C for 48 h. Equivalent weights (5 g) of the dried soil samples from each site were sieved and ground in an agate mortar and next were pooled (3 samples per pool; 5 pooled samples per site), and aliquots (~3 g) were digested with *aqua regia* under controlled condition (PN, 2002). A final determination of the total mercury content, both of fungi and soil substrate, was by cold-vapour atomic absorption spectroscopy (CV-AAS), using a fully

Fig. 1. Sampling sites (1–12; dots) of Brown Birch Scaber Stalk and certain main cities (circles) across Poland.

Table 1
Data on the total Hg measurement (μg/g dry matter) of certified materials and results of intercalibration trials

Standard material/trial	Certified value[a]	Measured value[a]
IAEA-359; cabbage leaves	13	11 ± 1
NIST-SERM 1570a; spinach leaves	30 ± 3	33 ± 1
CTA-OTL-1; oriental tobacco leaves	43[b]	41 ± 2
Aqacon Project 9 "Soil analysis"	0.3[c]	<1

[a] $n = 3$.
[b] Informative value.
[c] ng/g.

automated mercury monitor (Mercury monitor 3200, Thermo Separation Products, USA).

2.3. Analytical quality/analytical control

The method of mercury measurement was validated and controlled on several occasions by participation in international calibration activities, e.g. GESM/Food Euro proficiency testing exercise, IAEA trials, Aquacon Project 9 "Soil Analysis", and analysis of certified plant material (Table 1) and within-run reproducibility control (Falandysz & Chwir, 1997; Raport ICHTJ, 2002).

2.4. Soil pH and carbon

To a 10 g aliquot of air-dried soil sample, 50 ml of distilled water were added and the mixture was left for 1 h at 25 °C. Thereafter, pH was determined using a pH meter (EC20 pH/ISE Meter, Hatch) (Musgrove, 1987). Soil organic carbon content was determined gravimetrically after combustion of soil (10 g air-dried) organic matter at 800 °C in a furnace oven (CNOL 8,2/1100, Lithuania).

3. Results and discussion

3.1. Soil Hg

Topsoil collected from the stands where carpophores of *L. scabrum* were found was slightly acidified (pH from 3.54–4.38 to 5.85–6.24) at all twelve sites sampled, while organic carbon content varied, depending on the site, from 0.5–0.6 to 6.7–20.1% (Table 2). The total mercury content of top soil, after hot extraction with *aqua regia,* for most of the sites was within the range 0.026 ± 0.010 to 0.037 ± 0.014 μg/g dry matter. In topsoil from the Gostyńsko-Włocławskie Forest, County of Starachowice and the Lubelska Upland, in the central and south-eastern part of Poland, the total mercury content was 0.045 ± 0.016–0.066 ± 0.018 μg/g dm, which is greater ($p < 0.05$; U Man Whitney test) than other *L. scabrum* populations, including the southernmost, localized at the Kłodzka Hollow site in the Sudety Mountains (Table 2). These data did indicate a very low degree of forest soil pollution with mercury in Poland.

3.2. Brown Birch Scaber Stalk Hg

3.2.1. Concentrations

The caps (pileus) of Brown Birch Scaber Stalk, from most of the sites, exhibited total mercury at mean concentrations of 0.38 ± 0.23 to 0.64 ± 0.25 μg/g dm (median from 0.36 to 0.65 μg/g dm) and, for the Gostyńsko-Włocławskie Forest and the County of Starachowice sites these were 1.1 ± 0.4 and 1.2 ± 0.4 μg/g dm (median 1.2 and 1.1 μg/g dm, respectively) (Table 2). In the stalks (stem, stipe), total mercury content was lower than in caps, and for two sites in this study, which were the most contaminated, namely the Gostyńsko-Włocławskie Forest and the County of Starachowice, these were 0.52 ± 0.24 and 0.72 ± 0.20 μg/g dm (median 0.54 and 0.72 μg/g dm, respectively); other sites were 0.17 ± 0.08 to 0.35 ± 0.14 μg/g dm (median 0.17 and 0.33 μg/g dm, respectively). Overall-mean total mercury contents for 240 caps and stalks were, respectively, 0.63 ± 0.38 (0.072–2.0 μg/g dm) and 0.32 ± 0.20 (0.028–1.2 μg/g dm).

The total mercury content of carpophores of Brown Birch Scaber Stalk collected from most of the sites in Poland in 1989–1998 was between 0.054 ± 0.036 and 0.46 ± 0.33 μg/g dm for the caps and 0.044 ± 0.011 and 0.35 ± 0.14 μg/g dm for the stalks (Falandysz & Chwir, 1997; Falandysz, 2002; Falandysz & Kryszewski, 1996; Falandysz, Danisiewicz, & Gałecka, 1995; Falandysz, Marcinowicz, Danisiewicz, & Gałecka, 1997; Falandysz et al., 2002; Falandysz, Bielawski, Kawano, Brzostowski, & Chudzyński, 2002; Falandysz et al., 2003; Falandysz, Kawano, Świeczkowski, Brzostowski, & Dadej, 2003; Falandysz et al., 2004). At two of three other sites surveyed earlier, the total mercury content of caps and stalks of Brown Birch Scaber Stalk was somewhat elevated, e.g. 0.70 ± 0.27 and 0.35 ± 0.14 μg/g dm in the Łukta and Morąg Counties and 1.2 ± 0.7 and 1.1 ± 0.4 μg/g dm in the Borecka Forest (Falandysz, Gucia, Skwarzec, Frankowska, & Klawikowska, 2002; Falandysz et al., 2002). An elevated total mercury content was found in mushroom species collected from Trójmiejski Landscape Park in Gdańsk with a mean value of 6.7 ± 2.3 μg/g dm (median 7.3 and maximum 9.6 μg/g dm) in the caps and 4.6 ± 1.7 (median 4.6 and maximum 7.7 μg/g dm) in the stalks. Top soil total mercury content at this site was small, i.e. 0.010–0.095 μg/g dm (Falandysz et al., 2003). No statistically significant relationships could be noted between soil pH or organic carbon content and total mercury concentration of the caps or stalks of Brown Birch Scaber Stalk in the present study.

In several other studies in Europe, the total mercury content of carpophores of Brown Birch Scaber Stalk collected from background sites ranged from 0.19 to 0.72 μg/g dm (Aichberger, 1977; Allen & Steinnes, 1978; Cibulka et al., 1996; Kalač & Šlapetova, 1997; Kojo & Lodenius, 1989; Lodenius, Kuusi, Laaksovirta, Liukkonen-Lilja, & Piepponen, 1981; Svoboda, Zimmermannová, & Kalač, 2000). Exceptions to this pattern are specimens of

Table 2
Total mercury in cap and stalk of Brown Birch Scaber Stalk (*Leccinum scabrum*) and soil, and BCF values of mercury (μg/g dry matter; arithmetic mean, standard error, range and median value)

Site, year and number of specimens	Soil pH[b]	Soil C (%)[b]	Cap	Stalk	Soil	BCF_{Cap}	BCF_{Stalk}	Hg_C/Hg_S
1[a]. Darżlubska Forest, 2001 (15)[b]	4.85–6.24	4.6–12.9	0.42 ± 0.22	0.26 ± 0.17	0.030 ± 0.011	14 ± 4	8.1 ± 2.6	1.8 ± 0.6
			0.17–0.97	0.050–0.79	0.016–0.059	7.7–22	3.1–13	1.0–3.3
			0.36	0.24	0.32	15	7.9	1.6
2. Wdzydzki Landscape Park, 2000 (13)	4.76–5.43	2.4–3.6	0.46 ± 0.21	0.17 ± 0.08	0.033 ± 0.016	16 ± 10	6.0 ± 4.0	2.9 ± 1.0
			0.13–1.0	0.028–0.33	0.015–0.070	4.7–43	1.6–15	1.3–4.7
			0.41	0.17	0.030	15	4.5	2.7
3. Trójmiejski Landscape Park, 2000 (10)	4.31–4.82	4.8–12.5	0.51 ± 0.19	0.34 ± 0.20	0.036 ± 0.017	15 ± 2	9.3 ± 1.3	1.7 ± 0.5
			0.24–0.77	0.13–0.74	0.015–0.068	11–19	7.7–11	1.0–2.4
			0.59	0.29	0.032	15	9.2	1.7
4. Sobieszewska Island, Gdańsk, 2000 (13)	4.96–5.68	0.5–0.6	0.54 ± 0.26	0.24 ± 0.13	0.029 ± 0.011	19 ± 3	8.1 ± 1.7	2.4 ± 0.7
			0.23–1.1	0.12–0.62	0.016–0.051	14–23	5.8–12	1.4–4.0
			0.48	0.20	0.023	19	7.8	2.3
5. Tucholskie Forest, Osiek, 2001 (15)	4.52–5.48	3.4–11.2	0.59 ± 0.18	0.35 ± 0.14	0.036 ± 0.011	17 ± 3	9.7 ± 1.7	1.8 ± 0.6
			0.21–1.0	0.16–0.63	0.015–0.050	13–21	7.5–13	1.1–2.8
			0.59	0.33	0.037	16	9.7	1.8
6. County of Kętrzyn, 2000 (16)	4.52–4.94	5.1–18.2	0.43 ± 0.15	0.24 ± 0.09	0.034 ± 0.010	14 ± 6	8.1 ± 4.5	2.0 ± 0.9
			0.26–0.77	0.11–0.41	0.014–0.049	5.9–26	3.1–18	1.1–3.9
			0.39	0.23	0.035	14	7.1	1.6
7. Augustowska Forest, 1998–2001 (77)	3.80–5.23	5.2–17.4	0.65 ± 0.35	0.30 ± 0.17	0.036 ± 0.012	17 ± 6	8.2 ± 3.3	2.2 ± 0.7
			0.12–1.5	0.061–0.77	0.012–0.070	4.3–27	2.4–18	1.0–4.4
			0.65	0.30	0.036	18	8.3	2.0
8. Notecka Forest, 2000 (15)	4.78–5.29	5.4–10.8	0.64 ± 0.25	0.25 ± 0.11	0.037 ± 0.014	18 ± 3	6.7 ± 1.4	2.8 ± 1.1
			0.24–1.1	0.078–0.44	0.016–0.060	12–23	4.0–9.8	1.2–5.6
			0.63	0.28	0.043	18	6.6	2.7
9. Gostyńsko-Włocławskie Forest, 2001 (15)	4.54–5.61	2.6–15.3	1.1 ± 0.4	0.52 ± 0.20	0.056 ± 0.016	20 ± 4	9.2 ± 2.3	2.3 ± 0.8
			0.58–1.7	0.21–0.97	0.028–0.080	14–29	3.8–13	1.1–4.4
			1.2	0.54	0.057	21	9.4	2.1
10. Lubelska Upland, 2000–2001 (22)	4.53–4.98	2.2–7.8	0.58 ± 0.25	0.34 ± 0.17	0.045 ± 0.016	14 ± 5	7.9 ± 4.2	1.8 ± 0.4
			0.29–1.1	0.11–0.74	0.025–0.085	3.7–27	1.7–21	1.1–2.6
			0.53	0.30	0.043	13	6.8	1.7
11. County of Starachowice, 2001 (14)	3.76–4.90	6.7–20.1	1.2 ± 0.4	0.72 ± 0.20	0.066 ± 0.018	18 ± 2	11 ± 1	1.7 ± 0.4
			0.68–2.0	0.44–1.2	0.040–0.10	15–23	8.1–13	1.1–2.8
			1.1	0.72	0.064	18	11	1.6
12. Sudety Mountains, Kłodzka Hollow, 2000 (15)	3.54–4.38	5.6–15.8	0.38 ± 0.23	0.21 ± 0.08	0.026 ± 0.010	14 ± 5	8.0 ± 1.7	1.8 ± 0.8
			0.072–0.76	0.060–0.33	0.011–0.040	6.0–21	5.0–9.9	1.1–3.5
			0.36	0.23	0.026	12	8.3	1.3
All; 240			0.63 ± 0.35	0.32 ± 0.20	0.038 ± 0.016	16 ± 5	8.3 ± 3.1	2.1 ± 0.8
			0.072–2.0	0.028–1.2	0.011–0.10	3.7–43	1.6–21	1.0–5.6
			0.58	0.28	0.035	18	8.1	2.0

[a] For site location see Fig. 1.
[b] Number of samples (specimens).

Brown Birch Scaber Stalk collected from the mercury-polluted regions due to copper and mercury smelting in Slovakia, for which the total mercury concentration of carpophores ranged from 2.9 ± 3.0 to 15 ± 4 μg/g dm (1990–1993) (Kalač et al., 1996). Also, a highly elevated mercury content occurred in a specimen collected from an old mercury mine site in Germany with 6.2 μg/g dm in the cap (Fischer et al., 1995). These examples imply on bioindication ability of Brown Birch Scaber Stalk in regard to soil contamination with mercury, and this feature was confirmed for methylmercury (Fischer et al., 1995).

3.2.2. Hg BCFs and Hg_C/Hg_S

Based on the total mercury database available for Brown Birch Scaber Stalk (Table 2), this species can be classified as a moderate mercury accumulator. The bioconcentration factor (BCF) of mercury was calculated as quotient of the total Hg in cap/stalk to soil concentration. The BCF values of the total mercury in caps of Brown Birch Scaber Stalk from the particular sites ranged from 14 ± 5 to 20 ± 4 (total mean was 16 ± 5, and median 18) while, in stalks, they were from 6.0 ± 4.0 to 11 ± 1 (total mean was 8.3 ± 3.1, and median 8.1) (Table 2).

In several previous studies the BCF values of the total mercury in caps of Brown Birch Scaber Stalk ranged from 3.7 ± 4.2 to 40 ± 16, and in stalks were from 2.4 ± 2.4 to 40 ± 22 (Falandysz and Kryszewski, 1996; Falandysz, 2002; Falandysz and Chwir, 1997; Falandysz et al., 1995, 1997, 2001; Falandysz, Bielawski, Kannan et al., 2002; Falandysz, Gucia et al., 2002; Falandysz, Lipka et al., 2002, 2003; Falandysz et al., 2004). In one study, the total mercury BCF vales in caps and stalks of this species collected from the Trójmiejski Landscape Park in Gdańsk were especially high and reached on an average up to 150 ± 58 and 120 ± 100, respectively (Falandysz, Gucia et al., 2003).

The cap to stalk mercury concentration quotient for all 240 fruiting bodies of Brown Birch Scaber Stalk, was 2.1 ± 0.8 (median 2.0), while the range for this parameter was relatively narrow, i.e. from 1.7 ± 0.4 to 2.9 ± 1.0 for the particular sites (Table 2).

3.2.3. Hg intake

The Brown Birch Scaber Stalk total mercury data gained in this study for caps when converted to fresh weight (assuming 90% water content) on average were from 0.038 to 0.120 μg/g ww (total range from 0.0072 to 0.20 μg/g ww). A cap of this species is usually the most appreciated part of the carpophore, and also contains the total mercury at a greater concentration than that of stalk. The WHO limit for maximum weekly intake of total mercury is 300 μg (200 μg for CH_3Hg^+). Methylmercury usually forms only a small fraction of the total mercury in carpophore, 1.3% in Brown Birch Scaber Stalk, as reported by Fischer et al. (1995). Based on a "worst case" scenario possible, from this study (total mercury content 2.0 μg/g dm), weekly consumption of up to 1.5 kg fresh caps of Brown Birch Scaber Stalk originating from the background sites in Poland will not result in exceeding of the WHO limit for maximum weekly total mercury intake.

As mentioned in an introductory section, among the several sites surveyed in Poland earlier, highest mean, median and maximum mercury contents of 6.7 ± 2.3, 7.3 and 9.6 μg/g dm were recorded in specimens collected from the forested site in the city of Gdańsk. Usually the means of the total mercury content of the caps of Brown Birch Scaber Stalk in previous studies and this study were below 1.0 μg/g dm.

It is hard to assess the amount of wild mushrooms consumed *per capita* weekly by mushroom fanciers and people periodically dependent on or with easy access (villagers) to this kind of food, and not to mention exclusively Brown Birch Scaber Stalk intake rates. Based on human feeding, wild mushrooms, on average, are only a small part of the total diet, so consumption of Brown Birch Scaber Stalk collected from the background sites in Poland, or elsewhere in Europe, as regards mercury content, is safe.

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