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Total Mercury in Wild Mushrooms and Underlying Soil Substrate from the City of Umeå and Its Surroundings, Sweden

J. Falandysz,[1] M. Gucia,[1] A. Frankowska,[1] M. Kawano,[2] B. Skwarzec[3]

[1] Department of Environmental Chemistry and Ecotoxicology, University of Gdańsk, 18 Sobieskiego Str., PL 80-952 Gdańsk, Poland
[2] Department of Environment Conservation, Ehime University, 3-5-7 Tarumi, Matuyama 790-8566, Japan
[3] Department of Analytical Chemistry, University of Gdańsk, 18 Sobieskiego Str., PL 80-952 Gdańsk, Poland

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Mercury is one of the priority toxic metallic elements in the environment, which is ubiquitously dispersed and transported *via* the atmosphere on a global scale (Slemr *et al.* 1995). This element is one of the most toxic metals to humans and wildlife. Some mushroom species, especially those of the genus *Agaricus*, *Calocybe*, *Boletus*, *Lepiota*, *Lepista*, *Lycoperdon*, *Macrolepiota* have high bioaccumulation capacity for mercury (Lodenius and Herranen, 1981; Vetter and Berta, 1997; Kalač and Svoboda, 1998, 2000). The tolerance limit for mercury concentration in wild and cultivated mushroom in Czech Republic since 1999 is 5 and 1 μg/g dry weight, respectively (Kalač and Svoboda, 2000). Highly toxic methylmercury when present in food is usually considered as a compound well adsorbed by the gastrointestinal tract. The proportion of methylmercury to total mercury in the flesh of edible mushrooms is usually small, *i.e.* less than 20% (Stijve and Roschnik, 1974), while its real bioaviability from the mushroom diet by humans remains unknown. In practice nothing is known on the extent of leaching of inorganic mercury and methylmercury from the flesh of the edible mushrooms during processing and/or cooking.

In this study, species dependent differences/similarities in the bio-uptake efficiency of mercury were studied in some mushroom species in relation to the concentrations in soil near the city of Umeå and its surroundings.

MATERIALS AND METHODS

The fruiting bodies of 22 species of edible and inedible wild mushrooms and underlying soil substrate (Table 1) were collected from the forested areas of the city of Umeå (the suburbs of the Ersboda, Gamlia, Mariehem and area around of the Lake Nydala), and its surroundings (the villages of Ersmarck and Tavelsjö) in north-eastern Sweden in summer and autumn 1995. Only the fruiting bodies not infected with insects (larvae) were selected for the investigations.

The fruiting bodies immediately after pick-up were cleaned from the plant and substrate debris with a plastic knife, cut into small pieces, if necessary, and air dried for several days. The dried mushroom samples were packed in clean (fresh) polyethylene bags and kept in dry conditions until further dried in an laboratory oven at 40°C for 48 h, and then pulverised in agate mortar. The soil substrate samples (0-

Correspondence to: J. Falandysz

10 cm), after removal of the visible organisms, small stones, sticks, and leaves, were air dried at room temperature for approx. 4 weeks, and than sieved using a plastic sieve and ground in agate mortar and further dried in an oven at 40°C for 48 h.

For the determination of total mercury, the dried and usually pooled mushroom sub-samples (~300 mg) were wet digested in Teflon® vessels under pressure in an Automatic Microwave Digestion System (MLS 1200) using concentrated nitric acid Suprapur® "Merck". With every set a ~50 mushroom samples digested daily, two blank samples were prepared. Details of the analytical procedures are given elsewhere (Falandysz and Chwir 1997). Final determination was by cold-vapour atomic absorption spectrometry (CV-AAS) using a fully automated 3200 mercury monitor Thermo Separation Products, USA. The method was validated using biological reference materials, participation in inter-laboratory calibration trials and within-run reproducibility control (Falandysz, 1990).

RESULTS AND DISCUSSION

Concentrations of mercury, based on dry matter content, are given separately for the caps, stalks and/or whole fruiting bodies as well as substrate (Table 1). Table 1 also presents the bioconcentration factors (BCF) of mercury in mushrooms and the quotients of the concentration of mercury between caps and stalks. In addition, regression equations, correlation coefficients (r), and the confidence intervals of the linear relationship between mercury concentrations in the caps, stalks or whole fruiting bodies of the mushrooms and underlying substrate, as well as between BCFs of mercury and the degree of substrate contamination have been calculated.

Forest soil substrate rich in organic matter (decaying litter) contained mercury usually at concentration >100 ng/g dry matter, *i.e.* an order of magnitude higher than in soil mostly collected from the roads/pathways and road side (Table 1). The soil samples collected at the road side had the strata, somewhat disturbed due to human activities, and usually were poor in organic matter and often are rather sandy. The mercury deposition from the atmosphere onto terrestrial surface along the northern Swedish coast (including Västerbotten) is considered to be mainly due to past discharges, and the concentrations in soil are often in the range 250-400 ng/g dry wt (Informs, 1991; Rundgren *et al.* 1992). The measurements in 1995 have indicated that the median values of mercury in air of the trajectory between Berlin and Stockholm were of 1.93 to 1.54 ng/m^3. Both the concentrations and variability decreased from the most southern to the most northern site (Schmolke *et al.* 1999). The humus layer is thinner in soil in more northerly latitudes of Sweden and because of that even relatively small deposition rates of mercury can lead to continuous rise of its concentrations in soil, involving an increasing accumulation and risks due to toxic effects of mercury (Informs, 1991). The upper limit of mercury concentrations in humus rich soil substrate samples in this study is 450 ng/g dry wt and usually is below 380 ng/g dry wt, what is consistent with the data reported by other authors (Informs, 1991; Rundgren *et al.* 1992).

Table 1. The concentrations of total mercury (ng/g dry wt) in mushrooms and underlying substrate from the city of Umeå and its surroundings, bioconcentration factors (BAF) and Hg_C/Hg_S quotients of mercury

Species	No	Caps	Stalks	Substrate	BAF_{Cap}	BAF_{Stalk}	$Hg_C/_S$
False Morel *Gyromitra esculena*	1 (6)	22	31	450	0.049	0.069	0.71
Sheep Polypore *Albatrellus ovinus*	6 (30)	74±40 27-140	68±32 21-100	NA	NA	NA	1.1±0.2 0.80-1.4
Slippery Jack *Suillus luteus*	12 (48)	21±23 8.7-93	11±11 3.8-39	36±28 7.1-82	0.86±0.78 0.11-3.0	0.53±0.40 0.054-1.2	1.9±0.6 1.0-3.0
European Cow Bolete *Suillus bovinus*	6 (18)	66±53 11-130	33±29 4.5-70	64±54 11-150	0.90±0.51 0.42-1.7	0.42±0.18 0.14-0.62	2.2±0.7 1.2-3.1
Variegated Bolete *Suillus variegatus*	3 (9)	720±480 170-100	240±240 79-520	NA	NAp	NAp	4.1±3.6 1.9-8.3
Bay Bolete *Xerocomus subtomentosus*	3 (9)	150±40 120-190	110±20 87-130	NA	NAp	NAp	1.4±0.3 1.1-1.6
King Bolete *Boletus edulis*	16	1,200±1,400 58-5,400	610±910 19-3,700	55±65 3.1-240	25±20 0.92-52	12±11 0.18-34	2.6±1.0 1.3-5.2
Bitter Bolete *Tylopilus fellus*	1	140	86	360	0.39	0.24	1.6
Orange Birch Bolete *Leccinum versipelle*	13	500±310 100-1,200	300±230 57-940	140±110 53-360	6.1±5.2 0.52-17	2.8±1.7 0.42-5.8	1.9±0.6 1.1-3.3
Brown Birch Scaber Stalk *Leccinum scabrum*	19	180±130 33-470	100±60 15-210	80±58 24-190	3.7±4.2 0.33-12	2.4±2.4 0.17-6.4	1.7±0.8 0.74-3.7
Viscid Spike Cap *Gomphidius glutinosus*	10/8 (40/32)	42±21 11-68	35±26 12-89	54±52 14-190	1.1±1.0 0.28-3.4	0.74±0.46 0.41-1.7	1.9±1.8 0.40-5.7
Poison Pax *Paxillus involutus*	13 (39)	33±25 2.0-77	27±20 3.4-72	200±120 26-380	0.16±0.14 0.007-0.41	0.24±0.23 0.016-0.48	1.4±1.2 0.18-4.2

Sweating Mushroom *Clitocybe dealbata*	1 (20)	560	660	120	4.7	5.5	0.85
Anise-scented Clitocybe *Clitocybe odora*	1 (4)	1300	870	77	17	11	1.5
Sticky Gray Trich *Tricholoma portentosum*	1 (2)	1100	NA	27	41	NA	NA
Fly Agaric *Amanita muscaria*	16/15	390±540 9.4-1,900	220±300 5.0-990	66±44 15-170	3.8±4.1 0.20-11	2.2±2.7 0.11-7.9	2.0±1.0 1.4-5.2
Grisette *Amanita vaginata*	6	750±640 270-1,900	400±330 130-980	250±80 170-350	3.3±2.5 1.7-7.6	1.8±1.2 0.77-3.9	1.9±0.3 1.4-2.3
Russula *Russula badia*	8	13±11 1.9-32	6.1±1.9 3.8-9.0	230±50 160-270	0.072±0.066 0.0079-0.20	0.029±0.013 0.017-0.056	2.4±2.6 0.41-8.4
Cortinarius *Cortinarius hinnuleus*	4 (12)	1,900±1,500 400-3,700	1,400±700 340-2,000	250±70 170-290	11±6 3.9-15	5.8±3.0 2.4-8.2	1.8±0.1 1.2-1.8
Red-dappled Cort *Cortinarius bolaris*	1 (5)	4,200	2,000	710	5.9	2.8	2.1
Woolly Milk Cap *Lactarius torminosus*	8 (16)	23±10 9.2-39	12±6 4.3-21	28±9 7.4-37	1.2±1.7 0.30-5.3	0.70±0.94 0.14-2.8	2.0±0.5 1.0-2.4
Common Puffbal *Lycoperdon perlatum*	6 (30)*	1,600±700 770-2,800	NAp	100±130 18-250	65±29 11-110	NAp	NAp

Number of caps/numbers of stalks
In parentheses is given the number of caps/stalks per sample if the sample size was greater than one fruiting body
*A whole fruiting bodies
NA = Not analysed
NAp = Not applicable

There is an agreement that the process of mercury uptake from soil or litter by higher mushrooms is a complex phenomenon and for some species, it can be very efficient (Fischer *et al.* 1995). Common Puffbal, Sticky Gray Trich, Anise-scent Clitocybe and King Bolete were characterised by great (17-65) BCF of mercury in the caps. All these four species grew on soils containing small concentrations of Hg (mean below 100 ng/g dry wt), what implies/confirms that their high capacity to bioconcentrate mercury. An intermediate in its bioconcentration potential was Fly Agaric with a BCF of 3.8. Slipper Jack and European Cow Bolete, which grew on soils containing small concentrations of mercury accumulated lower concentrations of mercury (BCF below 1). Relatively great BCF values between 3.9 and 15 (mean 11) were recorded for Cortinarius collected from soil substrate containing between 170 and 290 (mean 250) ngHg/g dry wt., while for Grisette the BCF was 3.3 and soil mercury concentration was 250 ng/g dry wt. False morel, Poison Pax and Russula originating from mercury rich soil substrate (mean between 200 and 450 ng/g dry wt) were characterised by very small values (mean between 0.049 and 0.16) of BCF in the caps.

It is known that BCF of mercury in mushrooms exceeding 1 is usually found for the sites unpolluted with this element and an opposite situation (BCF <1) is observed for polluted sites. Near the cinnabar (HgS) mining and smelting plants, soil contained mercury concentrations ranging from 350,000 to 780,000 ngHg/g dry wt. (Bargagli and Baldi, 1984). Parasol Mushroom (*Macrolepiota procera*) very efficiently accumulated mercury ($p<0.001$; $r = 0.97$ for the caps and stalks, respectively) when this element was present in soil at concentrations between 37 and 93 ng/g dry wt. (Falandysz and Chwir, 1997).

The mercury content in the cap and stalk of European Cow Bolete increased with increasing concentration in soil ($p < 0.05$; $r = 0.83$ and 0.89, respectively), and the same was observed for the stalks of Viscid Spike Cap and whole fruiting bodies of Common Puffbal. On the other hand for the caps and stalks of Wooly Milk Cap and soil mercury concentrations a negative relationship was found ($p <0.05$; $r = -0.67$ and $r = -0.76$, respectively). Additionally, for the species such as Brown Birch Scaber Stalk, Orange Birch Bolete, Slippery Jack (stalks), Wooly Milk Cap and Common Puffball, the BCF values of mercury decreased ($0.01<p<0.05$) with increasing soil contamination.

A few countries have established legal tolerance limits for toxic metallic elements in edible mushrooms. The tolerance limit for mercury in Czech Republic since 1999, which is based on a daily intake rate of 300 g of fresh mushrooms, was 5 μg/g dry wt for wild edible species and 1 μg/g dry wt for cultivated species (Kalač and Svoboda, 2000).

Apart from the King Bolete, a most loved species by the pickers of wild edible mushroom, no other edible species contained mercury at the concentration exceeding value of 5 μg/g dry wt (Table 1). In the case of feral King Bolete, the concentration of mercury exceeded the mentioned value of tolerance in the cap of a single individual of the sixteen specimens examined, while mercury content of the large

Table 2. Total mercury concentrations in the mushroom species of the family *Boletaceae* Chev. from various geographical sites in Europe (ng/g dry weight)

Species, site and year	No	Cap		Stalk		Hgc/s	Ref.
King Bolete *Boletus edulis* Bull. ex Fr.							
Borecka Forest, Poland, 1998	16 (20)*	9,900±2,700	4,000-14,000	5,300±2,000	2,800-9,400	1.8	[1]
Szczybały Orłowskie, Poland, 1998	16 (27)	3,600±1,400	1,800-6,400	1,700±580	830-2,700	2.1	
Wdzydzki Landscape Park, PL, 1995-96	15	2,600±2,000	850-7,100	1,600±1,200	590-5,100	2.0	[2]
Umeå, Sweden, 1995	16	1,200±1,400	58-5,400	610±910	19-3,700	2.6	#
Commune of Gubin, Poland, 1994	16	3,000±1,200	1,200-4,500	1,500±600	380-2,600	2.0	[3]
Gdańsk area, Poland, 1989	16	97±27	64-170				[4]
Voivodeship Lubelskie, Poland, 1984-85	3	480	460-510	350		1.4	[5]
South Bohemia, 1994-96	8**	4,600±2,500	730-7,700				[6]
Copper smelter, Krompachy, 1990-93	5	32,000±19,000	up to 63,000				[7]
Mercury smelter, Rudnany, 1990-93	2	23,000	11,000-35,000				
Bohemia, unpolluted area, 1987-89	20	2,300±900	up to 4,400				[8]
Siena, Italy, p.1984	-	1,900					[9]
Mount Amita, Italy, p.1984	-	700					
Pinewood Bolete *Boletus aestivalis* Paulet *ex* Fr.							
South Bohemia, 1994-96	10**	3,000±3,000	300-8,600				[10]
Gdańsk area, Poland, 1989	15	470	130-1,800				[4]
Summer Bolete *Boletus pinophilus* Pil. *et* Derm.							
Province of Lugo, Spain, 1997	6¶	6,500±7,000	2,200-20,000				[11]
Gdańsk area, Poland, 1989	3	260±30	210-280				[4]

*Number of pooled samples and total number of the fruiting bodies (in parentheses); **a whole fruiting body; #this study; ¶without hymenophore (in hymenophore were 11,000±11,000 and range between 3,300 and 33,000 ng/g d. w.); [1] Falandysz *et al.* 2000; [2] Falandysz *et al.* 1999; [3] Falandysz & Kryszewski, 1996; [4] Falandysz *et al.* 1995; [5] Lasota & Witusik, 1987; [6] Kalač & Šlapetová, 1997; [7] Kalač *et al.* 1991; [8] Kalač *et al.* 1996; [9] Bargagli & Baldi, 1984; [10] Kalač & Šlapetová, 1997; [11] Alonso et al. 2000.

stalk of that species (~1 kg) was 3.7 μg/g. All seven specimens of King Bolete collected from the relatively sandy soil contained mercury at concentrations in the order of magnitude greater than nine specimens gathered from the forest soil rich in organic matter. Because of the methylmercury content of up to 20% of total mercury quantified in the flesh of the edible wild mushrooms and because of its bioavailability, excessive consumption of contaminated species may be a problem to the health.

Table 2 summarises some recent data on mercury concentrations quantified in the specimens of King Bolete, Pinewood Bolete and Summer Bolete, which are popular representatives of the *Boletaceae* family collected from various sites in Europe. Apparently, all the three species can bioaccumulate mercury to great concentrations. The degree of mercury contamination of the fruiting bodies of mushrooms growing at a contaminated site can be extraordinary high, and especially in King Bolete. Also the fruiting bodies of King Bolete originating from the background sites, which are located far away from industrial or urbanised areas such as the Borecka Forest in the Mazurian region in the north-eastern part of Poland (Table 2), can contain mercury at great concentrations. The cap to stalk mercury quotient for King Bolete is around 2 and is similar for the fruiting bodies collected from various spatially distant sites, and also with different history of pollution with metallic elements (Table 2).

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