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ORIGINAL ARTICLE

Soil-to-mushroom transfer and diversity in total mercury content in two edible Laccaria mushrooms

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Abstract Transfer factor and distribution of mercury were determined for *Laccaria amethystina* and *Laccaria laccata* growing in two distantly localized regions of the world and with different status of geogenic mercury. Mushrooms and soil were sampled in Poland in 2001–2013 and in the Yuxi region of Yunnan Province in China in 2013. The mushroom *L. laccata* was a more efficient accumulator of mercury than *L. amethystina*. Total mercury contents of whole fruiting bodies of *L. amethystina* and *L. laccata* at most sampling sites in Poland were low, i.e., around 0.02–0.1 mg kg^{-1} dry matter. For *L. laccata* from the Yuxi County of Yunnan province in China, Hg was at around 0.04 mg kg^{-1} dry matter. The median soil-to-mushroom transfer factors that are by some researchers also called bioconcentration factors calculated for *L. amethystina* were below 1, and for *L. laccata* they were from 3 to 20. The levels of contamination with mercury of both species do not pose toxicological problems, even if these species are consumed by mushroom fanciers in large amounts.

Keywords Bioconcentration · Mercury · Organic food · Wild-growing mushrooms

Introduction

Elemental mercury (Hg) easily diffuses into air and is prone to long-range atmospheric transport (Olivero-Verbel et al. 2015; UNEP 2013). Hg (>95 % as Hg°) in ambient atmosphere is slowly oxidized and undergoes deposition onto land and sea surfaces. Preferential deposition of oxidized Hg and Hg° on vegetation/air surfaces and soils was noted at higher elevations because of cloud and fog formation and high rates of rain or snow deposition (Ritchie et al. 2006; Schemenauer et al. 1995; Stankwitz et al. 2012; Zhang et al. 2013), and over time Hg accumulates in topsoil also by seasonal litterfall and throughfall (Demers et al. 2007). The process of Hg deposition is similar to that of persistent organic pollutants (POPs) by the "circumpolar wall" or cold condensation (Rappe 1974).

Mushrooms of different feeding strategies (saprobes, ectomycorrhizal) growing in background areas are often able to accumulate Hg and other metallic elements from the substratum by the live mycelium and sequester it into the fruiting bodies at high concentrations, e.g., Hg at much greater concentrations than other vegetation (Árvay et al. 2014; Falandysz et al. 2015a, b; Falandysz and Brzostowski 2007; Melgar et al. 2009; Nasr and Arp 2011; Saba et al. 2016; Tel et al. 2014). Hence, both edible and inedible mushrooms collected from uncontaminated areas often show Hg at much greater concentrations than other vegetation (Chudzyński et al. 2011; Falandysz et al. 2001a, 2013; Kojta et al. 2015; Krasińska and Falandysz 2015, 2016; Mleczek et al. 2015; Nasr et al. 2012; Rieder et al. 2011). Mushrooms with deeper mycelia in soils can absorb geogenic Hg at highly elevated concentrations at the sites with Hg geochemical anomaly due to occurrence of the global Circum-Pacific Mercuriferous Belt, and available examples showed that up to 4.8 mg kg^{-1} dry matter (dm) is in caps of edible Leccinum chromapes (Forst.) Sing in Yunnan, China (Falandysz et al. 2015b), up to 10 ± 3 mg kg^{-1} dm in caps of *Boletus aereus* in southern region of Spain and up to 13 mg kg^{-1} dm in this mushroom in Yunnan or up to 23 mg kg^{-1} dm in caps of *Boletus edulis* in Yunnan

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(Falandysz et al. 2015b, c; Ostos et al. 2015). A specific but locally important problem is occurrence of Hg at highly elevated concentrations in mushrooms growing in the sites more or less impacted by former Hg mines and smelters and other Hg deposits (Árvay et al. 2014; Kojta et al. 2015). These new data highlight a phenomenon of highly elevated concentrations of Hg noted in valuable for consumers' wild-growing mushrooms in regions of the world and exposure of consumers. In addition, mercury accumulated in fruiting bodies is largely retained in flesh during cooking and pickling, e.g., blanching (a short-time boiling of fresh mushrooms—typically for 10 min), and further is not lost in their pickling (Drewnowska 2015; Falandysz and Drewnowska 2015).

Litter, humus and soil solution are sources of minerals for mushrooms (Johanson et al. 2004). At places with elevated airborne Hg deposition due to long-range transport, litterfall can be a major pathway for Hg loading to humus and topsoil of forest catchments (Zhou et al. 2013). For mushrooms with shallow mycelia, contaminated litter seems to be a major source of Hg (Falandysz et al. 2014). This phenomenon has been well studied for radiocesium (134/137Cs) deposited from fallout onto surface of the forest floor, which is easily bioconcentrated by mushrooms with shallow mycelia such as *L. amethystina* (Falandysz and Borovička 2013; Falandysz et al. 2015a; Stijve and Poretti 1990).

This study is aimed at getting insight into the Hg accumulation by *Laccaria laccata* and *L. amethystina* collected individual foragers in Poland and elsewhere in Europe and also *L. laccata* widely collected across a whole China (Gumińska and Wojewoda 1985; Zhang et al. 2015; Mao 2009). Investigated was accumulation capacity of the element in the morphological parts of the fruiting bodies of *L. laccata* and *L. amethystina* and calculated were values of the Hg transfer factor in relation to levels in soil substrata. Also evaluated was intake of Hg from the *L. laccata* and *L. amethystina* by human consumers. *L. amethystina* and *L. laccata* are ectomycorrhizal species with shallow mycelia, which can be seen spreading in layers of decaying leafs in deciduous and coniferous forests largely in the Northern temperature zones. Both mushrooms largely depend on inorganic nutrients absorbed from litter. Both *L. laccata* and *L. amethystina* are common in Poland but are rarely foraged by individuals; in China, they are more popular and available at food markets.

Materials and methods

The Common Deceiver *Laccaria laccata* (Scop.) Cooke samples were collected in both Poland (several sites) and China (region of the Ligi and Jiuxi villages, near the city of Yuxi; N 24°21′/E 102°32′ in Yunnan in August 2013), and Amethyst Deceiver *Laccaria amethystina* (Bolt.: Hooker) MURR samples and soil substrates beneath were collected in Poland in 2001–2013 (Fig. 1). The fruiting bodies were separated into caps and stipes of which the bottom part was cut off to avoid contamination. The fungal materials and soils from certain sites were pooled to reduce costs of analyses, and each pooled sample contained from 8 to 97 specimens. The fungal materials were dried in 65 °C to constant weight and further were pulverized in a ceramic mortar and kept in new polyethylene bags under clean and dry conditions in depository room. The soils (0–10 cm layer) and litter samples free of any visible organisms, small stones, sticks and leaves were air-dried at room temperature for several weeks at free of dust conditions. Next, the soil samples were sieved through a 2-mm mesh plastic sieve and sealed in brand new polyethylene bags and kept under clean and dry conditions in depository room. Total mercury was determined using a direct sample matrices thermal decomposition and cold vapor atomic absorption spectroscopy (CV-AAS; MA-2000, Nippon Instruments Corporation, Takatsuki, Japan) (Brzostowski et al. 2011; Jarzyńska and Falandysz 2011).

The accuracy of the method was evaluated and further controlled by daily examination of fungal certified reference material (CS-M-1, dried fruiting bodies of *Suillus bovinus*, Institute of Nuclear Technology and Chemistry, Warsaw, Poland), with a nominal Hg concentration of 0.174 ± 0.018 mg kg^{-1} dry matter (dm), while our measurements showed 0.167 ± 0.004 ($n = 6$; average recovery at 96.0 %) and 0.165 ± 0.006 mg kg^{-1} dm ($n = 6$; average recovery at 94.8 %). For mushrooms and the soil substrates, the limit of detection was 0.0015 mg kg^{-1} dm, and the quantification limit was 0.005 mg kg^{-1} dm.

Results and discussion

Content of Hg in mushroom bodies

The range of values of Hg contents of *L. amethystina* and *L. laccata* of present study is listed in Table 1. For *L. amethystina*, the Hg median/mean contents of caps ranged from 0.011 to 0.15 mg kg^{-1} dm, and for stipes the values were usually lower (quotient of Hg content of caps and stipes ranged from 0.80 to 4.1; Table 1). For *L. laccata* from Poland, a range of Hg contents was even wider if compared to *L. amethystina*. The lowest median/mean value of Hg in caps of *L. laccata* was 0.022 mg kg^{-1} dm, around 30 times more was in specimens from the Włocławek forests, which showed 0.71 and for stipes was 0.57 mg kg^{-1} there. This is unknown what could be a local source of elevated Hg content determined in *L. laccata*

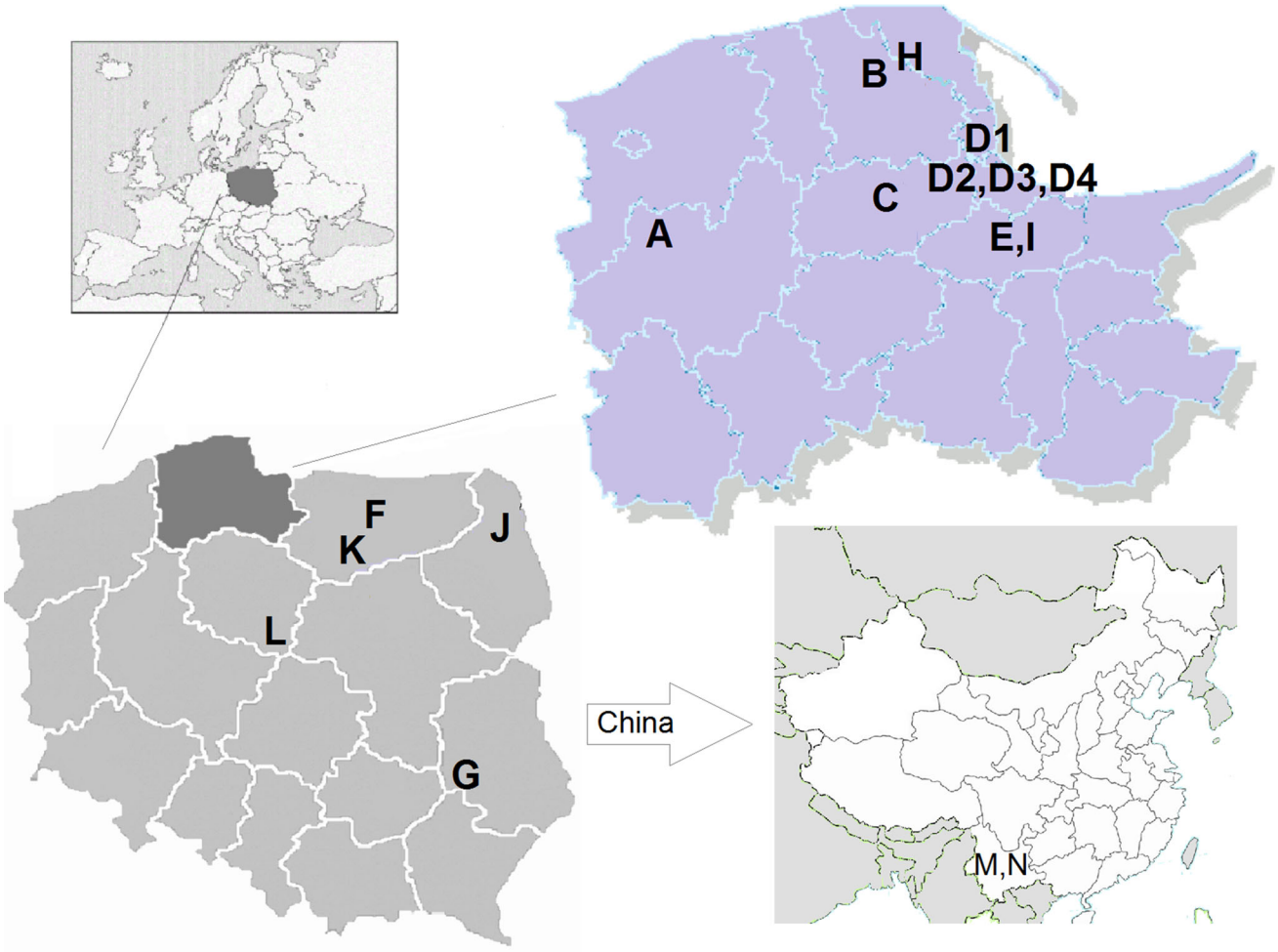

Fig. 1 Sampling sites in Poland (*A*, Dolina Słupi Landscape Park, N 54°30′/E 17°07′; *B* Commune of Łącze, N 54°63′/E 17°88′; *C* Mirachowo, N 54°40′/E 18°03′; *D1* Gdynia, N 54°50′/E 18°55′; *D2–D4* Gdańsk, N 54°35′/E 18°67′; *E* Jodłowno, N 54°25′/E 18°38′; *F* Orzechowo, N 53°57′/E 20°47′; *G* Poniatowa, N 51°83′/N 22°13′; *H* Puszcza Darżlubska, N 54°60′/E 18°23′; *I* Pomlewo, N 54°23′/E 18°37′; *J* Puszcza Augustowska, N 52°79′/E 23°43′; *K* Olsztynek, N 53°58′/E 20°30′; L, Włocławek, N 53°65′/E 19°03′) and China (M–N, N 24°21′/E 102°32′) (symbol of the place is explained in Table 1)

from the Włocławek forests. A possible explanation can be impacts from a nearby chemical industry, the Chemical Company Anwil S.A. This company was formerly known as the artificial fertilizers company: Zakłady Azotowe "Włocławek," and has been operated since 1966.

In addition to the Wloclawek site, an elevated content of Hg at 0.11–0.15 mg kg^{-1} also was in caps of *L. amethystina* sampled from the Trójmiejski Landscape Park at the Jaśkowa Dolina site, an area which has been heavily urbanized for over 100 years. Soils in the urban sites in Poland are more contaminated with Hg compared to background areas for which Hg content is <0.05 mg kg^{-1} dm (PIG 2013). In the forest ecosystem, both the litter and organic-rich surface soils can be affected by airborne Hg and other metallic element inputs from natural and anthropogenic, local and distant sources (Zhou et al. 2013).

The specimens of *L. laccata* collected from the region of County of Yuxi, Yunnan Province in China contained Hg at around 0.050 in caps and 0.030 mg kg^{-1} in stipes. Those values for *L. laccata* from Yunnan are similar to values determined for this species at several places in the northern part of Poland (Table 1).

There are few data published on Hg in *L. amethystina* from certain places in Europe, and one result is for *L. laccata*. In a cinnabar mining area in Tuscany (Italy), mushrooms and soil were collected and Hg in *L. amethystina* was at 0.64 and 1.0 mg kg^{-1} dm, while elevated was in soil beneath the fruiting bodies (4.4 and 35 mg Hg kg^{-1} dm in 0–4 cm layer, while the values of Hg TF were low, i.e., 0.14 and 0.03) (Bargagli and Baldi 1984). For regions uncontaminated with Hg, the data ranged for *L. amethystina* from 0.059 ± 0.030 to 0.067 ± 0.005 mg Hg kg^{-1} dm for two sites in Hungary (Vetter and Berta

Table 1 Mean, range and median values of Hg mg kg^{-1} dm content of caps and stipes of fruiting bodies of *Laccaria amethystina* and *Laccaria laccata* and beneath soil, and Hg cap to stipe concentration quotient ($Q_{C/S}$) and Hg bioconcentration factor (BCF) values

ID	Place, year and number of samples and specimens (in parentheses)	Hg			$Q_{C/S}$	BCF	
		Fruiting bodies		Soil		Fruiting bodies	
		Caps	Stipes			Caps	Stipes
Laccaria amethystina (Bolt.: Hooker) Murr.							
A	Pomerania, Commune Dębnica Kaszubska, Objezierze, Dolina Słupi Landscape Park, 2007 $n = 15$ (411)[a]	0.012 ± 0.004	0.0098 ± 0.0026	0.037 ± 0.014	1.3 ± 0.2	0.39 ± 0.24	0.31 ± 0.20
		0.0085–0.020	0.0061–0.013	0.010–0.062	0.77–1.6	0.18–1.1	0.14–0.93
		0.011	0.0093	0.018	1.2	0.32	0.25
B	Pomorskie Land, Wysokie, 2006 $n = 6$ (7)	0.043 ± 0.025	0.019 ± 0.006	0.054 ± 0.025	2.5 ± 2.1	1.1 ± 1.0	0.52 ± 0.49
		0.019–0.081	0.012–0.030	0.020–0.090	1.1–6.7	0.21–3.1	0.17–1.5
		0.038	0.018	0.054	1.8	0.73	0.34
C	Pomerania, Mirachowo, 2008 $n = 15$ (173)	0.023 ± 0.013	0.026 ± 0.009	0.098 ± 0.049	0.94 ± 0.54	0.28 ± 0.25	0.42 ± 0.40
		0.014–0.065	0.016–0.053	0.022–0.26	0.30–2.5	0.12–1.0	0.11–1.3
		0.019	0.025	0.12	0.81	0.20	0.20
D1	Pomerania, TLP[b], Gdynia, 2006 $n = 15$ (138)	0.015 ± 0.002	0.010 ± 0.001	0.090 ± 0.044	1.4 ± 0.1	0.21 ± 0.12	0.15 ± 0.08
		0.0092–0.017	0.0077–0.012	0.026–0.19	1.2–1.6	0.074–0.54	0.048–0.34
		0.015	0.011	0.083	1.4	0.19	0.14
D2	Pomerania, TLP, Dolina Radości, 2001 $n = 3$ (15)	0.033 ± 0.005	0.034 ± 0.006	0.11 ± 0.03	0.99 ± 0.22	0.32 ± 0.11	0.32 ± 0.07
		0.029–0.039	0.027–0.040	0.064–0.16	0.79–1.2	0.20–0.39	0.25–0.40
		0.033	0.036	0.10	0.97	0.38	0.31
D3	Pomerania, TLP, Gdańsk, Jaśkowa Dolina, 2001 $n = 3$ (9)	0.15 ± 0.01	0.11 ± 0.02	[c]WD	1.5 ± 0.1	WD	WD
D3	Pomerania, TLP, Gdańsk, Jaśkowa Dolina, 2001 $n = 1$ (8)[d]	0.13		0.16	WD	0.81	
D3	Pomerania, TLP, Gdańsk, Jaśkowa Dolina, 2001 $n = 3$ (12)	0.12 ± 0.03	0.12 ± 0.01	0.13 ± 0.04	0.80 ± 0.21	1.0 ± 0.3	1.3 ± 0.9
		0.084–0.15	0.11–0.13	0.065–0.16	0.65–0.95	0.69–1.3	0.72–2.0
		0.11	0.12	0.14	0.80	1.1	1.3
D4	Pomerania, TLP, Gdańsk, Niedźwiednik, 2013 $n = 1$ (97)	0.020	0.011	WD	2.1	WD	WD
E	Pomerania, Jodłowno, 2013 $n = 1$ (91)	0.12	0.029	WD	4.1	WD	WD
F	Warmia Land, Orzechowo, 2007 $n = 13$ (86)	0.017 ± 0.002	0.015 ± 0.002	0.046 ± 0.015	1.1 ± 0.1	0.40 ± 0.13	0.36 ± 0.12
		0.014–0.021	0.011–0.018	0.025–0.066	0.96–1.5	0.24–0.61	0.22–0.54
		0.017	0.016	0.043	1.1	0.41	0.36
G	Lubelskie Land, Poniatowa, 1999/2001 $n = 12$ (14)	0.10 ± 0.03	0.13 ± 0.05	0.11 ± 0.02	0.90 ± 0.48	0.91 ± 0.28	1.1 ± 0.42
		0.067–0.18	0.079–0.21	0.057–0.15	0.48–2.3	0.60–1.5	0.66–2.0
		0.095	0.099	0.12	0.81	0.85	1.0
Laccaria laccata (Scop.) Cooke							
H	Pomerania, Puszcza Darżlubska, 2003 $n = 15$ (191)	0.078 ± 0.017	0.066 ± 0.017	0.018 ± 0.001	1.2 ± 0.2	4.4 ± 1.0	3.8 ± 1.0
		0.051–0.11	0.041–0.11	0.016–0.019	0.96–1.4	2.7–6.4	2.1–6.1
		0.076	0.065	0.018	1.1	4.2	3.6
C	Pomerania, Mirachowo, 2008 $n = 1$ (29)	0.17	0.12	0.19	1.4	0.89	0.63
I	Pomerania, Pomlewo, 2013 $n = 1$ (20)	0.030	0.019	WD	1.6	WD	WD

Table 1 continued

ID	Place, year and number of samples and specimens (in parentheses)	Hg			$Q_{C/S}$	BCF	
		Fruiting bodies		Soil		Fruiting bodies	
		Caps	Stipes			Caps	Stipes
J	Augustów Land, Puszcza Augustowska, 2006 $n = 7$ (9)	0.12 ± 0.12	0.11 ± 0.13	0.031 ± 0.018	1.2 ± 0.3	6.2 ± 9.6	5.9 ± 9.1
		0.041–0.37	0.026–0.37	0.011–0.051	0.75–1.6	0.83–28	1.0–27
		0.056	0.051	0.034	1.2	2.9	2.0
K	Warmia land, Olsztynek, 2003 $n = 1$ (15)	0.022	0.025	WD	0.88	WD	WD
L	Kujawy Land, Włocławek forests, 2006 $n = 1$ (96)	0.71	0.57	0.074	1.2	9.6	7.7
M	Ligi, Yuxi, Yunnan, China, 2013 $n = 1$ (56)	0.054	0.029	WD	1.9	WD	WD
N	Jiuxi, Yuxi, Yunnan, China, 2013 $n = 1$ (71)	0.047	0.033	WD	1.4	WD	WD

[a] Pooled samples (number of specimens in parentheses)

[b] TLP (Trójmiejski Landscape Park)

[c] WD (without data)

[d] Pooled sample (if one pooled sample was made per place, it was examined in triplicate and the result is the mean value)

1997) and between 0.10 (0.08–0.11) mg kg^{-1} dm ($n = 6$) for a site in Germany (Seeger and Nützel 1976) and 0.74 ± 0.26 mg kg^{-1} dm in Turkey (Sesli and Tüzen 1999), while not provided was information on Hg content in soil substratum or values of TF.

The fruiting bodies of both species are edible, but their caps and stipes are small in size. Hence, the value of using the mushrooms for dietary purposes depends on dietary habit and is relatively low in Poland while they are more popular in Yunnan of China. In Poland, both *L. amethystina* and *L. laccata* are only sporadically collected by mushroomers. The values of Hg contents determined for *L. amethystina* and *L. laccata* for majority of the places in Poland were small, i.e., around 0.002–0.19 mg kg^{-1} if calculated for whole fruiting bodies on a fresh product basis (assuming moisture content at 90 %) (Zielińska et al. 2008; Falandysz et al. 2001b). Hence, the levels of contamination with Hg of both species do not possess a toxicological problem even if eaten occasionally by fanciers at greater volume or used as spices. Total Hg content of fresh whole fruiting bodies of *L. laccata* collected in the region of the city of Yuxi in the Yunnan province is around 0.004 mg kg^{-1}, which is also small value when compared to many other species of edible wild-grown mushrooms that are usually much more contaminated (Falandysz et al. 2007; Drewnowska et al. 2014; Gucia et al. 2012; Kojta et al. 2012).

The comparison of Hg content in analyzed species

The mushrooms *L. amethystina* and *L. laccata* and soils were collected from several distantly distributed sites, while both species were available for study only from the Mirachowo site. Content of Hg in caps and stipes of *L. laccata* at the Mirachowo place was at 0.17 and 0.12 mg kg^{-1} dm, respectively, and was several times greater compared to 0.019 and 0.025 mg kg^{-1} dm, respectively, for *L. amethystina* from this site. This discrepancy in Hg load between two species of mushrooms with similar feeding strategy and having shallow mycelia could be attributed to genetic features and availability of ligands to sequester Hg in fruiting bodies but less to environmental factors, which seem largely the same for both species.

The village of Mirachowo is inhabited by around eight hundred people and is surrounded by the large complex of Mirachowskie forests, which is part of the Kaszuby Landscape Park in the northwestern region of Poland (Fig. 1). There is no local industrial activity in the village of Mirachowo, while combustion of wood and less of hard coal seems to be a solely local source of Hg release into the atmosphere. Deposition of airborne Hg due to a long-range transport from the spatially distant sources is also possible, but rates are unknown for this area. Contents of Hg in surface layer of soils beneath the fruiting bodies for *L. amethystina* and *L. laccata* at this site were similar, i.e., 0.12 and 0.19 mg kg^{-1} dm (Table 1), while no litter was available for examination. Nevertheless, further study including collections of species litter, humus and mineral layer of soils from a several sites could give a deeper insight why both mushrooms differ in efficiency of Hg bioconcentration.

Soil-to-mushroom transfer factor (TF) is a value of quotient of Hg content of mushroom and soil. The median values of Hg TF for *L. amethystina* were usually below

unity and for *L. laccata* were higher, i.e., from 3 to 20, so the mushroom *L. laccata* is more efficient accumulator than *L. amethystina* (Table 1). This observation seems to support suggestion that *L. laccata* compared to *L. amethystina* could be more abundant in transporter genes and/or ligands are able to sequester Hg in fruiting bodies but evidence is lacking.

Some authors suggested that *L. amethystina* is species which can be suitable as indicator (early indicator) of an radioactive cesium ($^{134/137}$Cs) fallout (Byrne 1988; Stijve and Poretti 1990). This is because a shallow mycelia of *L. amethystina*, despite a moderate content of the stable (^{133}Cs) in fruiting bodies, allows for rapid uptake of the airborne $^{134/137}$Cs deposited on the forest floor through litterfall and throughfall. The mushroom *L. amethystina* is also known because of a high content of arsenic (As) that largely occurs in its fruiting bodies in the form of dimethylarsinic acid (Larsen et al. 1998; Zhang et al. 2015). This species when growing in soil contaminated with arsenic pentoxide (500–800 mg As kg^{-1} at a hot spot) accumulated in flesh total As at 1420 m kg^{-1} dm and largely biosynthesized in form of dimethylarsinic acid (68 %) (Larsen et al. 1998).

Conclusions

The results obtained in the study showed that mushroom *L. laccata* is a more efficient accumulator of Hg (calculated median values of transfer factor were from 3 to 20) than *L. amethystina* (values of the transfer factor are below 1). Mercury contents of whole fruiting bodies of *L. amethystina* and *L. laccata* for majority of the places in Poland were around 0.002 and 0.01 mg kg^{-1} fresh product, whereas for *L. laccata* from the Yuxi County of the Yunnan province in China was around 0.004 mg kg^{-1} fresh product. The levels of contamination with Hg of both species do not possess a toxicological problem even if eaten occasionally by fanciers at greater volume or used as spices.

The median values of Hg TF for *L. amethystina* were usually below unity and for *L. laccata* were higher, i.e., from 3 to 20, so the mushroom *L. laccata* is more efficient accumulator than *L. amethystina.*

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

Informed consent Informed consent was obtained from all individual participants included in the study.

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