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Review

Arsenic and arsenic speciation in mushrooms from China: A review

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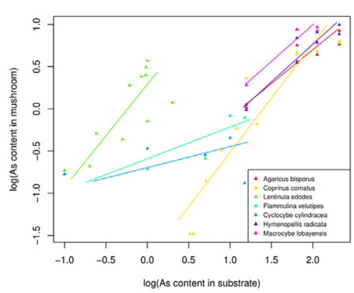
HIGHLIGHTS

- A review was carried out for studies on As and As species in mushrooms from China.
- As in mushrooms associated with environments and health risks is reviewed.
- Mushrooms possessing elevated As contents belong to several fungal families.
- Both intra- and inter-specific variation in As species in mushrooms can be large.
- Future perspectives for studies on As in mushrooms have been also discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) is a natural environmental contaminant to which humans are usually exposed in water, air, soil, and food. China is a typical high-As region, and also a great contributor of the world production of cultivated edible mushrooms and a region abundant in wild growing edible mushrooms. Mushrooms can accumulate different amounts of As and different As compounds, so potential health risk of As intake may exist to people who use mushrooms with elevated As contents as food or medicine. A systematic literature search was carried out for studies on As and As compounds in mushrooms from China. We compiled existing data from published sources in English or Chinese and provide an updated review of

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Organoarsenicals
Inorganic arsenic
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the findings on As in mushrooms associated with environments and health risks. Future perspectives for studies on As in mushrooms have also been discussed.

Contents

1. Introduction

Arsenic (As) is a natural environmental contaminant to which humans are usually exposed in water, air, soil, and food (Hughes et al., 2011). Acute effects of As poisoning include vomiting, abdominal pain, and diarrhoea, while chronic As exposure may cause various types of cancers. The toxicity of As greatly depends on its chemical form and oxidation state (speciation). The toxicity has conformed to the following order (highest to lowest): arsines > inorganic arsenites > organic trivalent compounds > inorganic arsenates > organic pentavalent compounds > arsonium compounds > elemental As (Komorowicz and Barałkiewicz, 2011). In general, amongst As compounds found in nature inorganic As is more toxic than organic As, while As compounds like arsenobetaine (AsB) and arsenocholine (AsC) are considered to be non-toxic. However, the phenylarsenic compounds and others, e.g., Lewisite, which have been produced and used as chemical warfare agents are extremely toxic (Leermakers et al., 2006).

Arsenic can be released from the lithosphere to the terrestrial soil by weathering of rocks, geothermal and volcanic activities release As (Zhu et al., 2014), however, the soil As content can be elevated by the anthropogenic sources of As (Adriano, 2001). Most of the anthropogenic As emissions can be accounted for ore smelting, coal combustion, and the use of As compounds in many products and production processes (Bissen and Frimmel, 2003; Finkelman et al., 1999; Matschullat, 2000). The anthropogenic atmospheric inputs (smelting and mining operations and fossil fuel utilization) play a role in the global redistribution of As, but the largest accumulations are generally in soils and/or sediments close to the source (Cullen and Reimer, 1989). Inorganic As in the forest floor soil can also be transferred to surface water by superficial flow under heavy rainfall (Huang and Matzner, 2007).

China is a typical high-As region, where 20 provinces have high As groundwater among 34 provinces (Guo et al., 2014). The average As content in top soil (A horizon) in China is 11.2 mg/kg which is more than twice the number of the world (Wei et al., 1991; Weng et al., 2000). Moreover, the As content in soil in Yunnan province is 18.4 mg/kg which is 1.6 times higher than its average content in China (Li and Wang, 2008). In addition, China is by far the world's largest producer and consumer of coal. The total As preserved reserves owned by Guangxi, Yunnan, and Hunan provinces accounted for 58% of the country's total reserves (Xiao et al., 2008). The As content in coals of Yunnan province can be significantly affected by the factors, such as the geochemistry of the source region and the tectonic and depositional environment of the basin and surrounding paleo-watershed (Zhou and Ren, 1992). In Yunnan, the discovery of wells, coal, and ore mines with high levels of As is continuing sporadically, and a similar scattered distribution pattern of patients is also being observed (Sun, 2004; Taylor et al., 1989; von Lindern et al., 2011).

Arsenic enters the human biosphere through microbial transformations (Zhu et al., 2014). Arsenic metabolism and cycling by microbes could take place 2.7 billion years ago (Sforna et al., 2014). Like other organisms, microbes take up As (V) in the form of arsenate via phosphate transporters and As (III) in the form of arsenite by aguaglyceroporins (Rosen, 2002). Fungi play an important role in As methylation (Gadd, 2007). Fungi are able to transform inorganic and organic As compounds into volatile methylarsines (Tamaki and Frankenberger, 1992). Arsenic accumulation and volatilisation by fungi rely on different processes, such as biomass sorption, complexation, oxidation/reduction, and biomethylation (Su et al., 2011), although demethylation of As in fungi may limit As volatilisation efficiency (Su et al., 2015). The major volatile compounds are mono-, di-, and trimethylarsine, respectively, and the major non-volatile compounds are methylarsonate and dimethylarsinate (Bentley and Chasteen, 2002). On the basis of chemical plausibility, the Challenger pathway with S-adenosylmethionine as a donor of CH_3^+ remains the most rational option to describe the biological methylation of As in organisms such as fungi and bacteria (Cullen, 2014; Kruger et al., 2013). Arsenobetaine formation is likely associated with the reproductive life stage of the *Agaricus* fungi (Nearing et al., 2016), since mycelium was not responsible for the biosynthesis of AsB under axenic conditions (Nearing et al., 2015).

Fungi can accumulate different amounts of As and different compounds of As in their fruiting bodies (mushrooms) mainly because of proteins that are present in fungi and resistant to As (Falandysz and Rizal, 2016; Shen et al., 2013), so the questions have been raised whether the edible mushrooms contain elevated As contents and what the compositions of As forms in different mushroom species are (Falandysz and Borovička, 2013). Increasing concern and fear also have been expressed to the presence of toxic elements (e.g. As) in Chinese herbal products which include medicinal mushrooms (Chan, 2003; Tripathy et al., 2015). Potential health risk of arsenicals intake may exist to people who use mushrooms with elevated As contents as food or medicine.

China contributed more than 70% of the world production of cultivated, edible mushrooms in 2016 (FAOSTAT, 2016). In 2016, the total mushroom production was nearly 36,000,000 t (http://www.cefa.org.cn/2017/10/24/10250.html). Therefore, China could be a great contributor to the improving of food production in terms of both quantity and quality (El Sheikha and Hu, 2018). Yunnan province in southwest China is at a transitional zone from tropical Southeast Asia to subtropical East Asia geographically. Therefore, it is a key area in biogeography and is one of the hotspot for biodiversity (Zhu et al., 2005), and is with biogeochemical anomalies of As in soils or sediments in the regions – such as Gejiu, Lincang, and Tengchong (Cheng et al., 2010; Dai et al., 2015; Zhu et al., 1986). Yunnan is also a specific region abundant in wild growing fungal species because of its geological and climate conditions (Wang et al., 2014). In Yunnan, nearly 900 fungal species are identified as edible (Wu et al., 2010). Many of the fungal species have been recorded as medicinal fungi, such as *Ganoderma lucidum*, *Tricholoma matsutake*, and *Wolfiporia cocos* (early named *Wolfiporia extensa*) (Dai and Yang, 2008; Wang et al., 2013).

Falandysz and Rizal (2016) reviewed As and its compounds in mushrooms. Two recent reviews on analytical methods of As species in mushrooms were also published (Braeuer and Goessler, 2019; Zou et al., 2019). However, many studies on As in mushrooms from China have been published in Chinese partly because of the language barriers. They are not fully accessible for those who do not read Chinese. Therefore, the aim of this review was to summarise the existing data on contents of As and As compounds in mushrooms from China and to provide an update review of the findings on As in mushrooms associated with environments and health risks.

2. Literature searching strategy

We performed a systematic literature search in ISI Web of Science and Google Scholar using combinations of key words including arsenic, fungi, mushroom, fruiting body, sporocarp, metalloid, China. Similar searching strategy was also carried out in CNKI (a Chinese literature database, http://www.cnki.com.cn/index.htm) in Chinese. We also checked cited references in the identified literature to find more relevant studies.

Contents (based on dry weight) of total As or As species were extracted from published studies, either from tables or from figures by WebPlotDigitizer (http://automeris.io/WebPlotDigitizer). The contents on wet weight were converted to the contents on dry weight based on the 90% moisture content in most of the fresh mushrooms. Bioconcentration factors (BCFs, content in mushroom/content in substrate) of As by mushrooms were calculated for the studies in which the As contents in substrate were also determined. BCF of As above 1 means that the mushroom actively concentrates As in its fruiting bodies. The dataset include the contents of As or As compounds from 87 studies which include 27 papers published in English and 60 papers or theses published in Chinese (Supplementary file 1 and 2).

Mushroom nomenclature followed the Index Fungorum (www.indexfungorum.org, accessed October 11, 2018). For mushrooms named as the trade names instead of the scientific names, we clarified the taxonomy of these mushrooms at genus or family level. Information for 164 mushroom species (82 genera from 39 families) was assembled including two phyla (*Ascomycota* and *Basidiomycota*). It should be noticed that phylogenetic analyses and DNA sequence comparisons have revealed that the species in China that named as "*Boletus edulis*" seemed to cluster more closely with *B. reticulatus* instead of the European and North American *Boletus edulis* (Feng et al., 2012). However, in this study we keep using "*Boletus edulis*" as the species name for analysing the data.

All data computations, statistical analysis, and graphics were performed using SPSS Statistics 22 (IBM, USA) or R 3.4.0 program (R Core Team, 2017).

3. Contents of total arsenic in mushrooms

In general, As in cultivated mushrooms were lower than in wild grown mushrooms. In cultivated mushrooms from China, the highest total As content was 42.3 mg/kg dw in a pooled *Lentinula edodes* sample purchased in a local supermarket in Kunming city, Yunnan province (Chen et al., 2017), but the value was from not detected (nd) to 9.9 mg/kg dw in the rest of the cultivated mushrooms. In wild grown mushrooms, the range of total As was nd to 212.3 mg/kg dw. The highest total As content in wild grown mushrooms was detected in a pooled sample called "black boletus" which was also purchased in Kunming city (Chen et al., 2017). The "black boletus" in that study could be *Boletus tomentipes*, *Retiboletus griseus* (formerly named *Boletus griseus*), *Sutorius brunneissimus* (formerly named *Boletus brunneissimus*), or other edible mushroom species from family *Boletaceae* with black color (Wang et al., 2004).

In phylum *Ascomycota* from China, relative high As contents were found in the mushrooms from families *Cordycipitaceae* and *Tuberaceae* (Table 1), such as *Cordyceps hawkesii* (4.9 mg/kg dw) from Anhui province (Sun et al., 2017), *Drechmeria gunnii* (7.6–12 mg/kg dw; 9.9 mg/kg dw on average) from Guizhou province (Sun et al., 2017), and *Ophiocordyceps sinensis* (0.42–13 mg/kg dw; 7.4 mg/kg dw on average) from Tibet autonomous region and Qinghai and Sichuan provinces (Chen, 2010; Falandysz and Rizal, 2016; Guo et al., 2018; Mleczek et al., 2018; Sun et al., 2017; Zhou et al., 2017, 2018), and *Tuber indicum* (12 mg/kg dw) from Yunnan Province (Liu et al., 2015).

In worldwide, as far as we know, the highest total As content in *Ascomycota* mushrooms (even in the kingdom Fungi) was 7100 mg/kg dw in *Sarcosphaera coronaria* (Borovička, 2004). Below 0.03 mg/kg dw As was detected in dried powder of *Ophiocordyceps sinensis* (formerly named *Cordyceps sinensis*) purchased from the pharmacy in Serbia (Stilinović et al., 2014). The value is much lower than that in *Ophiocordyceps* species collected in China. In *Morchella esculenta* from Germany, the As content was 0.32 ± 0.45 mg/kg dw (Rossbach et al., 2017), which is similar to the As content in *Morchella* species collected from China.

In phylum *Basidiomycota* from China, mushrooms possessed elevated As contents belong to several families (Table 1). For family *Agaricaceae*, by adding As to substrate, As contents were nd-8.4 mg/kg dw and <0.1–6.3 mg/kg dw in *Agaricus bisporus* and *Coprinus comatus*, respectively (Cai et al., 2011; Huang and Xu, 2014). In the studies without As adding experiments, elevated As contents (4.6–5.9 mg/kg dw) have been found in cultivated *Agaricus* spp. mushrooms purchased from markets in Shanghai city (Wang et al., 2016), whereas As contents were below 3.6 mg/kg dw in *Agaricaceae* mushrooms in other studies. For family *Boletaceae*, elevated As contents could be detected in the fruiting bodies of *Suillellus*

Table 1
Total arsenic contents in mushrooms from China in previous studies.

Family	*n*	Range of total arsenic content (mg/kg dw)
Phylum Ascomycota		
Cordycipitaceae	43	0.17–13
Morchellaceae	11	0.15–1.4
Pyronemataceae	1	0.15
Shiraiaceae	1	0.11
Tuberaceae	2	0.090–12
Phylum Basidiomycota		
Agaricaceae	65	nd-8.4
Albatrellaceae	1	0.27
Amanitaceae	4	0.080–1.4
Auriculariaceae	23	0.064–2.8
Bankeraceae	2	0.21–0.92
Boletaceae	232	nd-210
Cantharellaceae	8	nd-1.4
Cortinariaceae	1	0.25
Fomitopsidaceae	1	0.071
Ganodermataceae	29	0.010–1.4
Gomphaceae	6	0.15–4.0
Gomphidiaceae	2	0.25–0.29
Hericiaceae	3	0.10–0.25
Hydnangiaceae	16	0.075–160
Hymenochaetaceae	1	0.030
Hymenogastraceae	1	nd
Lyophyllaceae	9	0.18–9.1
Marasmiaceae	4	0.23–0.75
Meripilaceae	1	0.66
Mycenaceae	3	0.055–0.63
Omphalotaceae	126	0.019–42
Phallaceae	3	<0.05–1.8
Physalacriaceae	28	0.018–1.1
Pleurotaceae	71	nd-9.4
Pluteaceae	5	0.020–1.8
Polyporaceae	80	0.00070–2.6
Russulaceae	41	nd-12
Sclerodermataceae	3	0.16–1.7
Sparassidaceae	3	0.15–0.84
Strophariaceae	23	nd-1.2
Suillaceae	14	0.24–1.5
Thelephoraceae	3	0.15–44
Tremellaceae	14	0.024–4.4
Tricholomataceae	80	nd-22

luridus (120 mg/kg dw), *Leccinellum griseum* (17 mg/kg dw), *Boletus tomentipes* (13 mg/kg dw), *Sutorius magnificus* (13 mg/kg dw), *Boletus edulis* (13 mg/kg dw), *Butyriboletus roseoflavus* (7.8 mg/kg dw), *Boletus bicolor* (5.6 mg/kg dw), and *Xerocomus* sp. (5.6 mg/kg dw) (Falandysz et al., 2017c; Komorowicz et al., 2019; Xing et al., 2016; Yang et al., 2016; Zhang et al., 2019). For family *Hydnangiaceae*, genus *Laccaria* mushrooms, already known as As accumulators (Stijve et al., 1990), possessed 51 mg/kg dw As content on average (Duan et al., 2017; Liu et al., 2012; Zhang et al., 2005, 2015). For family *Lyophyllaceae*, As content was 0.18–9.1 mg/kg dw (3.6 mg/kg dw) in *Hypsizygus marmoreus* (Chen et al., 2017; Yang et al., 2013; Zhang et al., 2011, 2012). For family *Omphalotaceae*, all of the samples are *Lentinula edodes* mushrooms. Except one pooled sample purchased from a market in Yunnan province had 42 mg/kg dw As in *Lentinula edodes* fruiting bodies (Chen et al., 2017), As contents were no more than 3.7 mg/kg dw in the rest of the samples. For family *Pleurotaceae*, the highest As content was 9.4 mg/kg dw in a pooled sample of *Pleurotus citrinopileatus*, followed by 8.5 mg/kg dw in a pooled sample of *Pleurotus abalonus* (Chen et al., 2017), while the As contents were no more than 3.0 mg/kg dw in other *Pleurotus* mushrooms (species such as, *Pleurotus eryngii*, *Pleurotus geesterani*, and *Pleurotus ostreatus*). For family *Russulaceae*, the species with highest As content on average (7.6 mg/kg dw) is *Russula alutacea* (Liu et al., 2015). For family *Thelephoraceae*, 44 mg/kg dw was detected in *Thelephora vialis* from Yunnan Province (Yin et al., 2012). For family *Tricholomataceae*, the range of As contents was 0.97–9.9 mg/kg dw and 1.9–9.3 mg/kg dw in *Hymenopellis radicata* and *Macrocybe lobayensis*, respectively, by adding As to substrates (Huang and Xu, 2014). In the studies without As adding experiments, the fruiting bodies of *Tricholoma matsutake* (most of the samples were collected from Yunnan province) possessed 0.2–22 mg/kg dw (4.5 mg/kg dw on average) of As (Duan et al., 2017; Huang et al., 2010; Huang, 2011; Komorowicz et al., 2019; Liu et al., 2015; Yin et al., 2012; Zhang et al., 2004, 2012).

In recent years, several studies have been published on As in phylum *Basidiomycota* mushrooms from other countries. For example, Mleczek et al. (2015b) found As content was 1.6–2.8 mg/kg dw for *Boletus edulis*, 0.68–2.66 mg/kg dw for *Leccinum scabrum*, and 1.3–4.1 mg/kg dw for *Imleria badia* (formerly named *Boletus badius* or *Xerocomus badius*) in Poland. The As content was 0.54–0.70 mg/kg for caps and 0.36–0.46 mg/kg for stipes in *Boletus aereus* growing on volcanic and sedimentary soils in Italy (Alaimo et al., 2018). However, the highest As content in the *Imleria badia* mushrooms grown in extremely polluted waste and soil was 61–128 mg/kg dw (Mleczek et al., 2015a). In *Macrolepiota procera* from Serbia, As contents were nd-3.4 mg/kg dw and 0.20–2.8 mg/kg dw in the cap and the stipes, respectively, when the As content in soil was 6–25 mg/kg dw (Stefanović et al., 2016). In another study on *Macrolepiota procera* from Poland, slightly higher As content (0.37–5.4 mg/kg dw; 1.1 mg/kg dw on average) was detected in the caps (Falandysz et al., 2017b). Up to 17 mg/kg dw As has been found in *Tricholoma matsutake* from Korea (Choi et al., 2012). In other studies, relatively low As content found in *Sarcodon imbricatus* (1.0–1.9 mg/kg dw) and in *Amanita fulva* (0.28–0.78 mg/kg dw for caps and 0.25–1.3 mg/kg dw for stipes) from Poland (Falandysz et al., 2017a; Mędyk et al., 2017), and in *Pleurotus* species (0.51 mg/kg dw) from Bangladesh (Rashid et al., 2018).

4. Contents of arsenic species in mushrooms

Contents of As species in mushrooms from China have been investigated in several studies (Table 2). The extraction efficiency of As species was 55.3–104% from those studies (Supplementary file 1). Arsenic that was not extracted may be bound to lipids, cell components or proteins, or might even occur on the surface of the fungus as minerals (Koch et al., 2000). AsC has been only detected in Lentinula edodes at levels up to 0.011 mg/kg dw. The AsB content was 39 mg/kg dw in the Lentinula edodes sample with 42.3 mg/kg dw total As, but it was nd-0.085 mg/kg dw in other Lentinula edodes samples. For other mushroom species, the range of AsB contents was 0.0028–3.9 mg/kg dw. The highest content (200 mg/kg dw) of monomethylarsonic acid (MMA) was detected in the "black boletus" sample with 212.3 mg/kg dw total As, but the MMA contents were from nd to 51 mg/kg dw in the rest of the samples. The contents of dimethylarsinic acid (DMA) were from nd to 1.1 mg/kg dw in mushrooms from China, which was much less than the value (86 mg/kg fw; ca. 860 mg/kg dw) found in the "edible ink stain bolete", *Cyanoboletus pulverulentus* from the European countries (Braeuer et al., 2018c).

Arsenic (III) content in mushrooms from China was nd-2.0 mg/kg dw, while As (V) content was nd-6.2 mg/kg dw. The range of inorganic As (As (III) + As (V)) contents was 0.016 mg/kg dw in "black boletus" to 8.2 mg/kg dw in *Pleurotus citrinopileatus*. No research has been done on As species in *Laccaria* mushrooms from China, but DMA was found as the main As species (68%–86%) in *Laccaria amethystina* mushrooms collected from different European countries (Byrne et al., 1991; Larsen et al., 1998).

For some mushroom species, both the intraspecific and

Table 2
Contents of arsenic species in mushrooms from China in previous studies.

Family	AsC	AsB	MMA	DMA	As(III)	As(V)	iAs	Total As species	Total_As	Extraction efficiency	References
Ascomycota											
Cordycipitaceae											
Cordyceps hawkesii	–	0.073	nd	nd	0.13	1.0	–	–	4.9	–	Sun et al. (2017)
Drechmeria gunnii	–	0.030–0.068	nd	nd	0.16–0.22	0.88–2.3	–	–	7.6–12	–	Sun et al. (2017)
Ophiocordyceps sinensis	–	0.027–0.19	nd	nd	0.080–1.3	0.041–0.56	0.32–0.38	–	1.5–13	–	Guo et al. (2018) Sun et al. (2017) Zhou et al. (2018)
Basidiomycota											
Agaricaceae											
Agaricus blazei	–	–	–	–	0.624	0.264	0.58–0.89	3.2	0.83–3.6	88.9	González et al. (2009); Lin et al. (2012)
Agaricus spp.	–	–	–	–	–	–	0.12–0.37	1.9–2.9	2.2–5.9	86.4–90.6	Lin et al. (2012) Wang et al. (2016)
Boletaceae											
Boletus edulis	–	0.021–0.025	8.2	0.023–0.035	0.013–0.036	0.042–0.073	–	8.3–8.4	12–13	79–83	Komorowicz et al. (2019)
Suillellus_luridus	–	nd	37–51	0.070–0.086	nd	0.14–0.21	–	37–51	48–61	79–83	Komorowicz et al. (2019)
"Yellow boletus"	nd	0.18	nd	0.12	0.02	0.20	0.22	0.52	0.76	68.8	Chen et al. (2017)
"Black boletus"	nd	0.061	200	0.58	nd	0.016	0.016	200	210	93.9	Chen et al. (2017)
Cantharellaceae											
Cantharellus cibarius	nd	0.057	nd	0.47	0.12	0.24	0.35	0.88	1.03	85.3	Chen et al. (2017)
Hericiaceae											
Hericium erinaceus	nd	0.014	nd	0.062	0.011	0.048	0.059	0.14	0.13	101.5	Chen et al. (2017)
Lyophyllaceae											
Hypsizygus marmoreus	nd	0.034	0.82	0.092	1.3	5.4	6.7	7.7	8.0	95.5	Chen et al. (2017)
Omphalotaceae											
Lentinula edodes	nd-0.011	nd-39	nd-0.022	nd-1.1	nd-0.74	nd-0.42	0.020–2.5	0.021–40	0.031–42.3	55.3–104	Chen et al. (2017) Chen S et al. (2018a) Chen S et al. (2018b) Lin et al. (2012); Tang et al. (2015, 2017); Xu et al. (2016)
Physalacriaceae											
Armillaria mellea	nd	0.018	nd	0.020	0.0050	0.047	0.052	0.090	0.13	69.2	Chen et al. (2017)
Pleurotaceae											
Pleurotus abalonus	nd	0.066	1.3	0.11	1.3	5.5	6.7	8.2	8.53	96.5	Chen et al. (2017)
Pleurotus citrinopileatus	nd	0.046	0.67	0.082	2.0	6.2	8.2	9.0	9.36	95.9	Chen et al. (2017)
Pleurotus eryngii	–	–	–	–	0.12	0.12	0.24	–	0.32	–	González et al., 2009
Pleurotus sp.	nd	0.022	0.012	0.023	0.037	0.031	0.052	0.12	0.13	95.4	Chen et al. (2017)
Russulaceae											
Russula virescens	nd	0.016	nd	0.0075	0.016	0.12	0.13	0.16	0.17	92.9	Chen et al. (2017)
Strophariaceae											
Cyclocybe aegerita	nd	0.0028	nd	0.016	0.041	1.1	1.2	1.2	1.2	102.6	Chen et al. (2017)
Pholiota sp.	nd	0.0041–0.059	nd	0.013–0.077	nd-0.11	0.063–0.079	0.075–0.19	0.13–0.21	0.16–0.25	79.4–87.5	Chen et al. (2017)
Tremellaceae											
Tremella sp.	nd	0.0038	nd	nd	0.019	0.022	0.041	0.044	0.043	102.3	Chen et al. (2017)
Tricholomataceae											
Lepista nuda	–	–	–	–	0.29	nd	0.29	–	0.29	–	González et al., 2009
Leucopaxillus giganteus	–	–	–	–	0.42	0.38	0.80	–	1.4	–	González et al., 2009
Tricholoma matsutake	–	2.8–3.9	0.61–1.1	0.039–0.087	nd	0.056–0.069	–	3.5–5.2	5.5–6.7	79–83	Komorowicz et al. (2019)

interspecific variation of As species can be large (Dembitsky and Rezanka, 2003; Komorowicz et al., 2019; Seyfferth et al., 2016). It has been previously confirmed that the major As compound in different As-accumulating mushrooms could be different (Byrne et al., 1995). Different As species profiles have been also found in three fungal species from As smelter sites in Austria (Kuehnelt et al., 1997). Nearing et al. (2014a) found differences of As speciation profiles among different fruiting body morphologies. In China, Chen et al. (2017) found arsenobetaine was the main As species in *Lentinula edodes* mushrooms. However, others found that inorganic As was the main As species in *Lentinula edodes* mushrooms (Chen et al., 2018b; Xu et al., 2016), which was similar to that of Llorente-Mirandes et al. (2014). In an investigation of wild grown mushrooms from Chongqing city, the ratio of AsB content to organic As content was 25% for bolete mushrooms, 83.8% for *Tricholoma matsutake*, and 60.4% for *Lactarius* sp., respectively (Gan et al., 2017).

5. Arsenic uptake in mushrooms

BCFs of As varied among fungal taxonomic groups (Fig. 1). The BCFs above 1 were found in the following families: *Amanitaceae* (genus *Amanita*), *Boletaceae* (genera *Boletus*, *Gyroporus*, *Pulveroboletus*, *Retiboletus*, and *Suillellus*), *Gomphidiaceae* (genus *Chroogomphus*), *Strophariaceae* (genus *Cyclocybe*), *Physalacriaceae* (genus *Flammulina*), *Hydnangiaceae* (genus *Laccaria*), *Russulaceae* (genera *Lactarius* and *Russula*), *Omphalotaceae* (genus *Lentinula*), *Sclerodermataceae* (genus *Scleroderma*), *Suillaceae* (genus *Suillus*), *Lyophyllaceae* (genus *Termitomyces*), *Tricholomataceae* (genus *Tricholoma*), and *Tuberaceae* (genus *Tuber*). Up to now, the highest BCF of As in mushrooms from China was 29 in the caps of *Laccaria vinaceoavellanea* collected from Yunnan (Zhang et al., 2015).

Arsenic adding experiments were carried out to investigate accumulation ability of several common edible commercial species (Fig. 2). The As contents in *Flammulina velutipes* and *Cyclocybe cylindracea* were not correlated with the As contents in their substrates ($p > 0.05$). However, the As contents in *Agaricus bisporus*, *Coprinus comatus*, *Lentinula edodes*, *Hymenopellis radicata*, and *Macrocybe lobayensis* increased with the As contents rising in the substrates. For these species, it is important to make sure low substrate As content in mushroom cultivation (Mleczek et al., 2016b).

For wild growing mushrooms, the toxic element contamination of these species in Yunnan becomes a critical issue due to elevated soil toxic element contents discovered in recent years. The ranges of As contents in the main soil types in Yunnan are 0.50–310, 0.10–630, and 1.2–180 mg/kg dw for larteritc, red, and yellow soil, respectively (Weng et al., 1997). The average soil As content in Yunnan is relatively very high (18–20 mg/kg dry weight, dw), compared with many other places in China (Weng et al., 1997). Average As content in coal from Yunnan was 22 mg/kg dw which is much higher than the average As content (9.7 mg/kg dw) in coal in China (Kang et al., 2011). In polluted soil of a Zn-Pb mining region in Yunnan, the main sources of As are mining activities, airborne particulates from smelters, and the weathering of tailings (Cheng et al., 2018).

Previous studies on wild growing mushrooms have shown that variations of elements between caps and stipes of *Boletus* fungi are

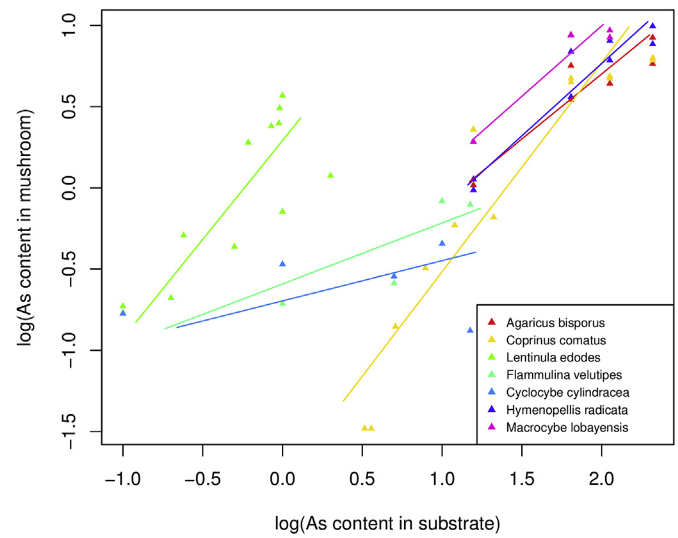

Fig. 2. Correlations of arsenic contents in mushrooms and in underlying substrates from arsenic adding experiments in China. Note: No certified reference materials have been used in these studies. *Agaricus bisporus*: y = −0.900 + 0.800x, r = 0.805, $p < 0.01$. *Coprinus comatus*: y = −1.803 + 1.287x, r = 0.836, $p < 0.01$. *Lentinula edodes*: y = 0.291 + 1.271x, r = 0.618, $p < 0.01$ *Flammulina velutipes*: y = −0.591 + 0.375x, r = 0.710, $p < 0.072$. *Cyclocybe cylindracea*: y = −0.696 + 0.247x, r = 0.035, $p < 0.765$. *Hymenopellis radicata*: y = −1.016 + 0.891x, r = 0.907, $p < 0.01$. *Macrocybe lobayensis*: y = −0.739 + 0.868x, r = 0.927, $p < 0.01$.

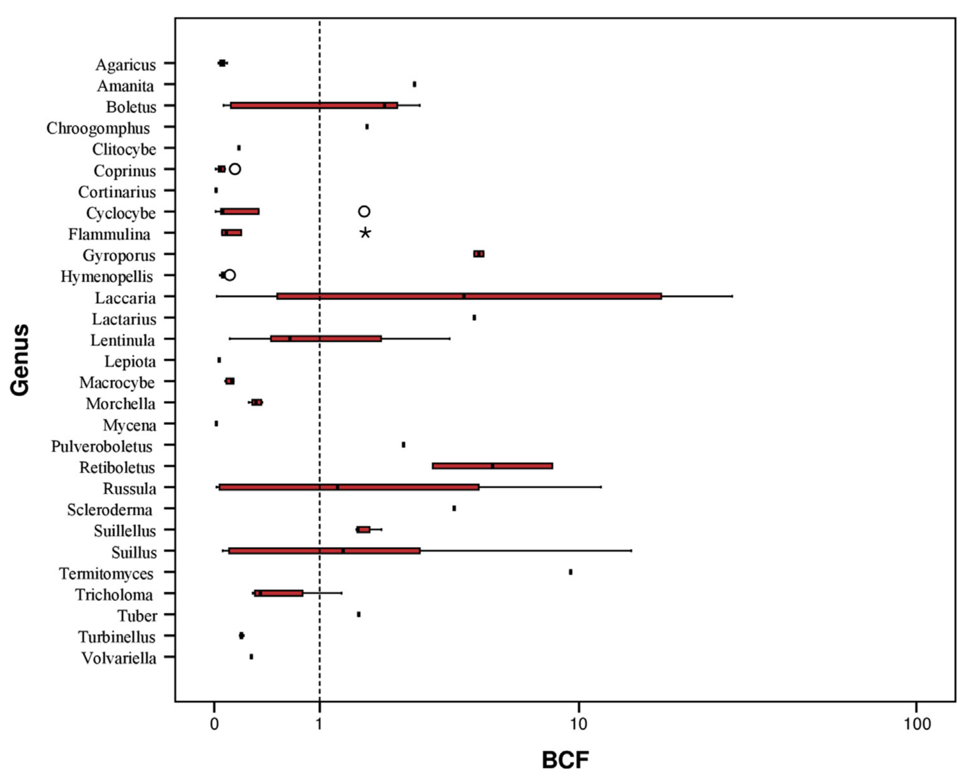

Fig. 1. Bioconcentration factors of arsenic in mushrooms from China in previous studies.

mainly related to different bedrock soil geochemistry, enrichment capability for various elements as well as mushroom species (Wang et al., 2015a, 2015b, 2017). The mean values of As contents in composite samples of caps for *Suillellus luridus*, *Sutorius magnificus*, and *Boletus tomentipes* from three to four locations in Yunnan were at the range of 0.79−53 mg/kg dw, therefore, *Boletus* spp. have the potential as bio-indicator of As geochemical anomaly (Falandysz et al., 2017c). Arsenic has been detected at level 450 mg/kg dw in *Imleria badia* (family *Boletaceae*) growing in close proximity of sludge deposits contaminated with this metalloid at level 490 mg/kg dw (Mleczek et al., 2016a). High levels (up to 27.1, 40.5 and 88.3 mg/kg dw for As (III), As (V) and DMAA, respectively) of As were also found in *Imleria badia* collected from a severely contaminated places in Poland (Niedzielski et al., 2013). In the Norwegian, Finnish, and Russian border regions, the Ni-Cu smelter is the main source of As in local wild foods from, and As content in the mushrooms was higher than that in other wild foods (Hansen et al., 2017).

6. Health risk assessment of arsenic in mushrooms

Health risk assessments of As in mushrooms from China have been carried out in numerous studies. In many of them, the total As contents in mushrooms were compared with the maximum permitted levels from different standards. According to the National Standard for Food Safety in China - Limits of Pollutants in Foods (GB 2762-2017), the maximum permitted levels of total As in edible plants and fungi is 0.5 mg/kg fresh weight (fw). In the standard for green food - edible fungi (NY/T 749-2012) established by the Ministry of Agriculture of China, the maximum permitted levels of total As is 0.5 mg/kg fw or 1.0 mg/kg dw (except 1.5 mg/kg for *Tremella fuciformis*). For medicinal mushrooms, the maximum permitted level for As is 4.0 mg/kg dw in the ISO 18664: 2015 Traditional Chinese Medicine - Determination of heavy metals in herbal medicines used in Traditional Chinese Medicine. However, no standard in China has been set for As species levels in mushrooms as far as we know. The Joint FAO/WHO Expert Committee on Food Additives (JEFCA) recommended that the provisional tolerable intakes of inorganic As were 3.0 μg/kg body weight (bw) per day (JECFA, 2010). For dimethylarsinic acid (DMA), the United States Environmental Protection Agency established tolerable daily intakes of 0.43 mg/kg bw (US EPA, 2006).

In other studies on As in mushrooms from China, the most popular way to investigate the As health risk assessment is comparing the As intake from mushroom meals with the limit dose for inorganic As established by the JECFA. When the estimated As intake from a certain edible mushroom species exceeded the limit dose, the mushroom was not recommended to consume. If there were only total As data, authors usually considered the total As content as the inorganic As content to calculate the As intake. In these cases, carcinogenic effect of As by eating mushrooms could be overestimated since inorganic As was not the only As species occurred in mushrooms.

7. Future perspectives

7.1. Advanced methods for arsenic determination

Elemental speciation analysis became a very useful tool in environmental monitoring, food quality control, and human health risk assessment (Marcinkowska and Barałkiewicz, 2016). HPLC coupled with ICP-MS, although expensive, is the most popular tool for element speciation in recent years. Methods based on HG-AAS are easier to implement and less costly than those based on HPLC-ICP-MS in terms of the analytical instrumentation needed (Fiamegkos et al., 2016). There is a potential of the HG technique coupled to ICP-OES for the non-chromatographic As speciation (Welna and Pohl, 2017). However, the application of HG-AAS or HG-AFS is not suitable for determining non-hydride-forming As species, such as AsB (Braeuer and Goessler, 2019). The use of additional analytical methods (such as X-ray absorption spectroscopy (XAS) and electrospray mass spectrometry (ESI−MS)) in a complementary manner introduces the ability to greatly enhance the information obtained from HPLC-ICP-MS analysis (Nearing et al., 2014b). The laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) is another advanced analytical method that allows direct measurement of the contents of As or other elements in a solid sample (Sajnóg et al., 2018). In a recent study on cultivated market mushrooms, by using LA-ICP-MS, Li et al. (2019) found that As in *Lentinula edodes* distributed to the surface coat of the cap, whereas As in *Pleurotus ostreatus* distributed to the junction of the pileus and stipe.

7.2. Detection of more arsenic species in more mushroom species

Recently, the occurrence of trimethylarsine oxide and methylarsonous acid at significant contents in *Elaphomyces* spp. has been reported (Braeuer et al., 2018a). Homoarsenocholine, a novel As compound has been detected for the first time in fungal species (Braeuer et al., 2018b). However, there is still a lot to be understood about the profile of As in mushrooms from China. The major components of As species in *Ophiocordyceps sinensis* were a cluster of unknown organic As compounds with As (III), which accounted for 91.7−94.0% of the total As content (Guo et al., 2018).

Mushroom samples from the smelter-polluted environment contained higher concentrations of unknown arsenicals and may thus represent perspective material for speciation studies (Borovička et al., 2019). The profile of As compounds in more mushroom species collected from more sites should be investigated. Šlejkovec et al. (1997) gave a perspective on understanding the pattern of As compounds in a wider range of genera of mushrooms and the relationships between fungal taxonomy or evolutionary status and the As compounds present. It is still an interesting area for further studies. In this case, a perfect mushroom identification and its status within classification using molecular methods are needed.

7.3. Effect of processing on arsenic in mushrooms

Very few studies have been reported on the effects of processing on As contents in mushrooms from China. The total As content in dried *Lentinula edodes* mushrooms decreased 39% after soaking for 60 min, and total As content in fresh *Lentinula edodes* mushrooms decreased 13% after blanching for 2.5 min (Xu et al., 2016). Chen et al. (2018a) found that approximately 3.4%−43% total As contents were released from *Lentinula edodes* samples into the blanching water after various microwave blanching. In two surveys on heavy metals pollution of commercial mushrooms in Shanghai city, the As contents (0.040 mg/kg fw and 0.12 mg/kg fw, respectively) in canned mushrooms were lower than those (0.053 mg/kg fw and 0.13 mg/kg fw, respectively) in fresh mushrooms (Bai et al., 2018; Xu et al., 2018). Study from Spain showed that the total As content decreased 53%, 59%, and 71% in *Agaricus bisporus*, *Lentinula edodes*, and *Pleurotus ostreatus*, respectively, after boiling for 10 min (Llorente-Mirandes et al., 2016). Drewnowska et al. (2017) found that blanching for 15 min could reduce by 86% (85−86%) content of total As (total As) in the caps of *Amanita fulva* Fr., while a further pickling (with vinegar) of blanched caps had little effect and total loss of As has been assessed as 89% (88−89%), when calculated on a dry weight basis. Another study also confirmed that As content in

Agaricus bisporus decreased significantly during industrial marinating process (Pankavec et al., 2019). Evidences from one more study suggested that cooking (a type condition not provided) could significantly, i.e. by 26–72% reduce As content in mushrooms – calculated on dry to dry basis (Chiocchetti et al., 2020), however, our knowledge is still poor on effects of processing on As speciation. In a very recent study by Chen et al. (2020), the transformations of some As species were observed in dried *Lentinus edodes* and *Agaricus blazei* mushrooms during ultrasonic treatment.

In Yunnan, one of the traditional ways to cook mushrooms is stir-frying mushroom slices ca. 5 min without pre-blanching. However, stir-fried mushrooms may contain lower concentrations of toxic elements (e.g. Hg and As) than fresh mushrooms when expressed on a dry basis, but the values may be higher on a whole (wet) weight basis (Falandysz et al., 2019a and 2019b; Unpublished, JF). Up to now, no available data on the change of As species content in mushrooms by this cooking method. In one study on the influence of different cooking processes on the bioaccessibility of As in two commercial fish, the As bioaccessibility slightly decreased after frying for 5 min (He et al., 2010).

7.4. Bioaccessibility of arsenic

The global interest in As speciation has continued in recent years with numerous publications, including studies on human exposure to dietary inorganic As and other As species (Clough et al., 2018). In the human body As is transported by the blood to different organs, mainly in the form of monomethylarsonous acid (MMA) after ingestion and MMA was more cytotoxic to human cells compared to inorganic As (III) and As (V) (Mandal and Suzuki, 2002). Therefore, As methylation may not only be a detoxification process but may also enhance toxicity and/or carcinogenesis to human (Duker et al., 2005). Since some organic As compounds and their metabolites can produce cytotoxic effects (Taylor et al., 2017), a full assessment of As (including organic As compounds) in mushrooms is needed to fill the current gaps in knowledge. It is necessary to measure bioaccessibility, together with As speciation, in foods such as mushrooms (Koch et al., 2013).

Chen et al. (2018a) used an *in vitro* physiologically based extraction test to evaluate the As risk assessment associated with *Lentinus edodes* intake. They found that, in gastric fraction and gastrointestinal fraction, the contents of As (III), DMA, MMA in mushrooms decreased, while As (V) content were increased, which implied that oxidation of As (III) and demethylation of DMA and MMA occurred. In another study, the bioaccessibility of As was 64.46% in whole *Ophiocordyceps sinensis*, 66.15% in caterpillar body, and 30.18% in stroma (Zhou et al., 2018).

Also, the authors Komorowicz et al. (2019) recently studied bioaccessibility of As compounds from powdered *B. bainiugan* (earlier described as *B. edulis*), *Tricholoma matsutake* and *Suillellus luridus* in gastric and gastrointestinal juices using enzymatic assisted extraction *in vitro*. The authors determined that bioaccessibility values were in the ranges from 73 to 102% (the mixture of the artificial solution of saliva) 74–115% (the mixture of the gastric juice) and 18–87% (the mixture of the gastric intestinal juice and bile).

8. Conclusion

In China As in cultivated mushrooms is lower than in wild grown mushrooms in general. The mushrooms possessing elevated As contents belong to several families from phyla *Ascomycota* and *Basidiomycota*, including families *Amanitaceae*, *Boletaceae*, *Gomphidiaceae*, *Strophariaceae*, *Physalacriaceae*, *Hydnangiaceae*, *Russulaceae*, *Omphalotaceae*, *Sclerodermataceae*, *Suillaceae*, *Lyophyllaceae*, *Tricholomataceae*, and *Tuberaceae*. For some mushroom species, both the intraspecific and interspecific variation in As forms can be large. Health risk assessments of As in mushrooms have been carried out in numerous studies by comparing the total As contents with maximum permitted levels from Chinese national or international standards or comparing the estimated As intakes with the limit dose recommended by JEFCA. Further work need to be done on the following: developing simpler methods for As compounds determination, identifying As species in mushroom species not investigated so far, identifying As in places of geochemical anomalies and As compounds in mushrooms there, investigating the effects of processing on As in mushrooms, and assessing the As health risk. The places with geochemical anomalies resulting in high levels of As in edible and medicinal mushrooms of China need to be further identified and characterized.

Declaration of competing interest

The authors declare no competing interests.

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Appendix A. Supplementary data

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