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# Bioaccumulation of Methylmercury versus Inorganic Mercury in Rice (*Oryza sativa* L.) Grain

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Mercury (Hg) bioaccumulation in aquatic food webs has been much studied, motivated from high Hg levels found in many fish species important for human consumption. Hg bioaccumulation in terrestrial food chains have received little attention and assumed to be of minor importance. However, recent studies showed that rice can be an important pathway of methylmercury (MeHg) exposure to inhabitants in Hg mining areas in China. In this study, 59 sampling sites (including 32 sites from “heavily polluted area”, 19 from “less-impacted area” and 8 from “control sites”) were selected in a Hg mining area in China and both inorganic Hg (IHg) and MeHg were determined in rice grain (brown rice) and soil samples to evaluate Hg bioaccumulation in rice grain. Bio-Accumulation Factors (BAFs) for IHg ranged from 0.00014 to 0.51 and from 0.71 to 50 for MeHg. BAFs for MeHg were on average more than 800 times higher than those for IHg (maximum: 40 000 times). This study, for the first time, showed that rice grain is an intensive bioaccumulator of MeHg, but not of IHg, which may be trapped by the roots.

## Introduction

Mercury (Hg) is a global pollutant, still receiving much attention decades after it was put on the environmental agenda (1). Organic compounds of Hg, such as methylmercury (MeHg), are of greatest concern due to the high toxicity and potentially severe effects on humans (2). MeHg can be generated from inorganic Hg (IHg) in the environment by micro-organisms under special conditions (3). MeHg is known to bioaccumulate strongly in the aquatic food chain, resulting in tissue concentrations in certain fish species more than  $10^6$  times higher than ambient water Hg concentrations (4). Food consumption is the main pathway for MeHg exposure to humans and the average absorption rate of MeHg by human body is about 95% (5). Tragic pollution incidents, such as in Minamata in Japan have highlighted the extreme

toxicity of MeHg and the importance of fish consumption as an exposure pathway (6). More recently, epidemiological studies have shown dose–effect relationships at much lower exposure (7–10).

Tremendous effort has been devoted to understanding the mechanism of MeHg bioaccumulation in the aquatic food webs in North America and Europe because fish consumption is generally believed to be the main exposure pathway of MeHg to humans. However, elevated concentrations of total Hg (THg) as well as MeHg in rice have been reported from Hg contaminated areas (e.g., MeHg reach  $144 \mu\text{g}\cdot\text{kg}^{-1}$  (11) and  $174 \mu\text{g}\cdot\text{kg}^{-1}$  (12)). In such contaminated areas, rice grains have been observed with high ratios of MeHg to THg (e.g., average as 45% (11)). Moreover, Feng et al. suggested that rice consumption is the major MeHg exposure pathway for a population in a Hg mining area in Guizhou, China (13).

Rice is the main food staple of more than half the world's population, particularly in Asia where more than 2 billion people get up to 70% of their daily dietary energy from rice and its byproduct (14). The few studies available on Hg in rice are mostly descriptive of concentrations only. The information on bioaccumulation characteristics of rice to different Hg species is still largely unknown. The present study aims at presenting for the first time, the bioaccumulation properties of MeHg and IHg in rice in a Hg polluted area with Hg contamination gradients from highly contaminated to lowly impacted. This study provides new evidence about MeHg accumulation in rice, and potential mechanisms of MeHg and IHg bioaccumulation in the rice-soil system.

## Materials and Methods

**Study Area.** The Wanshan area is a typical hilly and karstic terrain with elevations ranging from 205 to 1149 m above sea level. The region has a subtropical humid climate characterized by abundant precipitation and mild temperature. The annual average rainfall is 1200–1400 mm, and the annual mean temperature is 17 °C.

The Wanshan area was one of the major Hg producing regions of the world. The largest mine ranked third in the world (15). The region has five major river valleys (Supporting Information (SI) Figure S1), and all have or have had considerable Hg mining and retorting activities. Large scale Hg production activities ceased in 2001, but small scale artisanal Hg production were still operated in one of the valleys (Gouxu) at the time of sampling (2007), mainly using previously mined low grade ore. A large number (around 50) of very simple furnaces were observed along the banks of the upper parts of the river. In the other four valleys (Aozhai, Xiaxi, Gaolou, and Huangdao) Hg mining and retorting were abandoned in 2001.

The rivers are between 22 and 34 km long from upstream to where they join with one of the larger rivers Jinjiang River to the north or Wushui River to the south (SI Figure S1). Most Hg mines and smelters are situated in the upstream part of the rivers. Rice paddies are located throughout the valleys, from just downstream of the calcine piles to side valleys and tributaries. Hence there are strong gradients in Hg influence within a limited area, making the region particularly suitable for case studies of the environmental impacts of Hg contamination from Hg mining and retorting activities. Parts of the rice paddies are irrigated by river water contaminated by Hg from the mine tailings, if available rainfall or spring water from the nearby mountains or other tributaries are insufficient. More detailed information on the local environmental setting is available elsewhere (12, 13, 16).

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**Sample Collection and Preparation.** The sampling region (covering about 700 km<sup>2</sup>; SI Figure S1) was operationally divided into “seriously contaminated area” (32 sampling locations; with distance <8 km from the pollution sources), “less-impacted area” (19 sampling locations; with distance ≥8 km from pollution source) and “control sites” (the farthest eight locations from pollution source; those sites were not included in “less-impacted area”), respectively, to scrutinize the regional difference (sampling locations were shown in SI Figure S1). This division was based on a previous observation that the serious Hg contamination in river water with THg ≥50 ng·L<sup>-1</sup> (the U.S. EPA standard limit for fresh water) was limited to about 8 km from the pollution sources (17).

Rice grain samples were directly collected from paddy fields during late August 2007 (early harvest period), and soil samples were collected from the rice roots (about 5–10 cm top depth). Four typical sites (A9, A20, D4, D12; SI Figure S1) were selected to collect the whole rice plant to preliminarily scrutinize Hg distributions in different parts of rice (grain, leaf, stem, root, and hull). At each sampling site, a composite sample was composed of five subsamples. All rice and soil samples were individually sealed into three successive polyethylene bags avoid cross-contamination and transferred to the laboratory on the same day of sampling (soil samples were stored and transferred with ice packs).

In the laboratory, two subsamples of wet soil were first homogenized with a blender after removing bigger particles (e.g., stones and plant residues), and the first was discarded and the second were sealed in polyethylene bags and stored in freezer before analysis (within 28 days). Homogenization started with the control samples, followed by less contaminated and continued with more contaminated samples. The blender was carefully cleaned, first using tap water, then acid-washed, and then rigorously rinsed by ultrapure water and dried by a hair drier before a new sample cycle. Before measurement, each wet soil sample was divided in two, with one subsample for the determination of Hg, and the other for drying at 45 °C for moisture content determination.

As sampling were conducted at the early stage of the harvest period, the moisture content in soil samples were still elevated, with a mean of 47% ± 8% (ranging: 32–63%), which suggest that the paddy soil were not totally drained and impacts from demethylation on MeHg data due to aeration would not greatly underestimate the MeHg concentrations in the paddy soil.

Rice grain samples were washed with ultrapure water at least three times and dried at 40 °C until constant weight, then processed by first removing the hull from the seeds, and then the seeds were crushed and ground by a grinder (for whole rice plant samples, after separating the seed, other parts were divided into four fractions, including hull, root, stem, and leaf.). All precautions were taken in order to avoid any cross-contamination during the process. Two subsamples were ground, the first was discarded and the second powdered sample was subsequently sealed in a polyethylene bag and stored in a refrigerator. The grinder was rigorously rinsed by ultrapure water and dried by a hair drier before a new sample cycle.

**Analytical Method.** For THg analysis, soil samples were digested in a water bath (95 °C) using a fresh mixture of HCl (12 mol·L<sup>-1</sup>) and HNO<sub>3</sub> (16 mol·L<sup>-1</sup>) (1:3, v/v) (18), whereas rice samples were digested with a mixture of HNO<sub>3</sub> (16 mol·L<sup>-1</sup>) and H<sub>2</sub>SO<sub>4</sub> (18.4 mol·L<sup>-1</sup>) (4:1, v/v) (13). THg for soil samples was measured using cold vapor atomic absorption spectrometry (CVAAS), while THg for rice samples was determined using dual-stage gold amalgamation method and cold vapor atomic fluorescence spectrometry (CVAFS) detection following method 1631 (19). For MeHg analysis, rice samples were prepared using KOH-methanol/solvent ex-

traction (20), whereas soil samples were prepared using CuSO<sub>4</sub>-methanol/solvent extraction (21). For both, MeHg was determined using aqueous ethylation, purge, trap, and GC-CVAFS detection (22) following method 1630 (23). Soil pH was measured using a pH electrode and the solid: water ratio was 1:2.5 (24). Organic matter (OM) contents of soil samples were determined using the potassium dichromate volumetric method coupled with water heating (25). Inorganic Hg (IHg) was obtained by subtracting MeHg from THg. Measurements of total gaseous mercury (TGM) in ambient air were performed using a portable Zeeman Mercury Analyzer RA-915<sup>+</sup>. The analyzer operation is based on differential atomic absorption spectroscopy using high frequency modulation of the polarization of light. The detection limit of the instrument is 2 ng·m<sup>-3</sup> at the applied flow rate through the instrument of 20 L·min<sup>-1</sup>. Information on QA/QC of our measurement data is available in the Supporting Information.

## Results and Discussion

**Hg Levels in Rice.** Elevated Hg concentrations were found in the rice grain (brown rice), with mean (and range) of IHg concentrations in rice grain samples of 94 (7.4–460) μg·kg<sup>-1</sup> in the “seriously contaminated area”, 45 (6.0–170) μg·kg<sup>-1</sup> in the “less-impacted area”, and 25 (8–66) μg·kg<sup>-1</sup> at the “control sites”, respectively. Corresponding MeHg concentrations were 11 (1.2–44), 5.8 (1.4–24), and 4.7 (1.6–13) μg·kg<sup>-1</sup> in the three groups, respectively. IHg and MeHg in rice grain in the “less-impacted area” and the “control sites” were significantly lower ( $P < 0.05$  for all) than those in “seriously contaminated area”. The highest concentration in rice grain samples for each valley was generally observed just downstream of the mine tailings (so did the Hg in paddy soil) (SI Figure S2). Hg concentrations in rice grain in “seriously contaminated area” are close to previously reported values in the same area (11–13). Hg concentrations in rice at the “control sites” were close to a previously reported values for rice samples collected from 15 provinces in China (Shi et al. (26) have reported 19 (6.2–36) μg·kg<sup>-1</sup> for IHg and 4.5 (1.9–10.5) μg·kg<sup>-1</sup> for MeHg).

Approximately 90% of the samples from the “seriously contaminated area”, 60% of the samples from the “less-impacted area”, and 50% of the samples from “control sites” had rice grain concentration of THg above 20 μg·kg<sup>-1</sup>, recommended as the maximum value for consumption by the Standardization Administration of the People’s Republic of China (27). Furthermore, MeHg constituted a large proportion of THg in rice grain, with a mean (and range) of 17% (3.5–40%), 21% (3.3–40%), and 18% (6.9–40%) in the three groups, respectively. Four out of the total 59 samples had MeHg concentrations above 20 μg·kg<sup>-1</sup>, the limit for THg, clearly indicating potential health risk for local residents.

**Hg in Rice Paddy soil.** Mean (and range) of IHg in rice paddy soil samples was 35 (0.29–311) mg·kg<sup>-1</sup> in the “seriously contaminated area”, 5.9 (0.21–21) mg·kg<sup>-1</sup> in the “less-impacted area” and 0.86 (0.22–1.5) mg·kg<sup>-1</sup> at the “control sites”. Corresponding MeHg concentrations were 2.7 (0.27–12), 0.99 (0.14–2.4), and 0.78 (0.22–1.5) μg·kg<sup>-1</sup> in the three groups. Both IHg and MeHg concentrations in paddy soil in the “less-impacted area” and the “control sites” were significantly lower than those in the “seriously contaminated area” ( $P < 0.01$  for IHg and  $P < 0.05$  for MeHg). The concentrations in the “seriously contaminated area” were comparable to other reports from the same area: 0.10–790 mg·kg<sup>-1</sup> for IHg and 0.10–4.0 μg·kg<sup>-1</sup> for MeHg (16); 8.1–160 μg·kg<sup>-1</sup> for IHg and 1.3–23 μg·kg<sup>-1</sup> for MeHg (11). THg concentrations in paddy soils at the “control sites” was higher than that reported for uncontaminated soils worldwide (0.01–0.5 mg·kg<sup>-1</sup>) (28). Higher concentrations in this region may be expected due to elevated concentrations in the

**TABLE 1. Mercury Levels in Paddy Soil (also OM and pH) and in Rice Grain, MeHg Ratios to THg (M/T), and Bio-Accumulation Factor (BAF) in Wanshan Hg Mining Area (THg = IHg + MeHg)**

unit	paddy soil (dry weight)				pH	rice grain (dry weight)			BAF	
	IHg mg·kg <sup>-1</sup>	MeHg μg·kg <sup>-1</sup>	M/T %	OM %		IHg μg·kg <sup>-1</sup>	MeHg μg·kg <sup>-1</sup>	M/T %	IHg	MeHg
seriously contaminated area (<8 km from pollution source), (n = 32)										
mean	34.6	2.72	0.039	3.98	6.71	94.1	11.3	17.3	0.0041	5.61
SD	57.2	2.61	0.075	1.06	0.35	118	10.7	10.5	0.093	10.3
min	0.29	0.27	0.0014	1.95	6.13	7.44	1.19	3.46	0.00014	0.71
max	311	12.1	0.35	6.45	7.32	464	44.3	39.8	0.51	49.7
less-impacted area (≥8 km from pollution source), (n = 19)										
mean	5.89	0.99	0.076	2.51	6.79	44.6	5.78	20.8	0.013	6.88
SD	5.92	0.54	0.11	1.05	0.27	47.1	6.22	11.5	0.0095	7.37
min	0.21	0.14	0.0010	0.71	6.34	6.00	1.40	3.30	0.0010	1.60
max	21.1	2.41	0.43	4.45	7.39	167	24.0	39.8	0.030	30.5
control sites (the furthest sites from pollution source), (n = 8)										
mean	0.86	0.78	0.19	3.41	7.00	25.1	4.66	18.3	0.032	4.44
SD	0.42	0.48	0.14	1.38	0.31	19.8	3.54	10.1	0.019	2.98
min	0.22	0.23	0.044	2.49	6.52	8.00	1.60	6.90	0.011	0.90
max	1.48	1.57	0.49	6.12	7.34	66.0	12.6	35.5	0.057	10.6

bedrock in the area which was situated in the center of the circum-Pacific mercuriferous belt (15, 29) and elevated atmospheric Hg deposition (30).

For 85% of the paddy soil samples in the “seriously contaminated area” and 70% in the less-impacted area, THg concentrations exceeded the Chinese national standard limit for paddy soils (1.5 mg·kg<sup>-1</sup>) (31), showing widespread contamination to paddy soils from mining and smelting activities. From the control sites, all samples had concentrations below (or equal to) the limit value.

**Relationships between Hg in Rice and Hg in Soil.** The IHg concentrations in rice grain were significantly ( $P < 0.001$ ) lower than those in the corresponding paddy soils. On the contrary, the MeHg concentrations in rice grain were significantly ( $P < 0.001$ ) higher than the concentrations in corresponding paddy soil (Table 1). Mean (and range) site-specific bioaccumulation factors (BAFs, i.e. rice/soil concentration ratios (32)) for IHg was 0.0041 (0.00014–0.51) in the “seriously contaminated area”, 0.013 (0.0010–0.030) in the “less-impacted area” and 0.032 (0.011–0.057) at the “control sites”. Corresponding BAF for MeHg was 5.6 (0.71–50), 6.9 (1.6–31) and 4.4 (0.9–11), respectively, in the three groups. Hence, the BAF of MeHg is on average more than 800 times than those of IHg (maximum: 40 000 times) (Figure 1, Table 1). On average, the BAF of MeHg in the “seriously contaminated area” was similar to that in the “less-impacted area” and at the “control sites”, but BAF of IHg in the “seriously contaminated area” seem to be lower than

that in the “less-impacted area” and at the “control sites”, although this finding was not statistically significant ( $P > 0.05$ ) (Table 1).

There was no clear relationship between BAF of MeHg and BAF of IHg ( $P > 0.05$ ) (Figure 1), suggesting MeHg may have different accumulation mechanisms compared with IHg in the rice grain, as previously reported for other plants (4, 33).

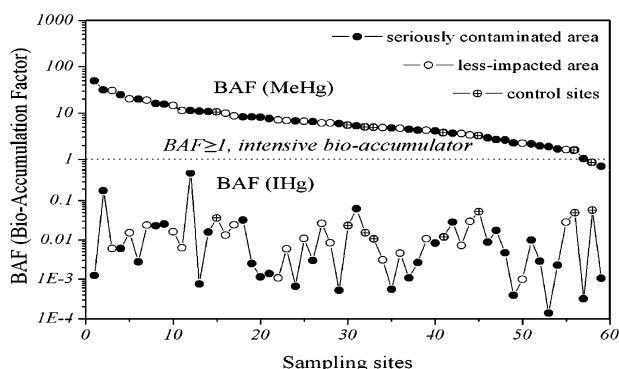
For comparison, we calculated the BAFs for the data reported by Horvat et al. (11) in the Wanshan Hg mining area (same as this study) and the Qingzhen area (also in Guizhou province, but without Hg mining). The BAF was 0.0022 (0.000083–0.013) for IHg and 9.0 (1.5–34) for MeHg ( $n = 10$ ) in Wanshan (same as the “seriously contaminated area”) and 0.0044 (0.00002–0.011) for IHg and 0.82 (0.2–1.1) for MeHg ( $n = 4$ ) in Qingzhen area, which are in good agreement with our results.

Higher accumulation of MeHg than of IHg in other plants has been previously reported (32). The BAF of IHg in rice grain is similar to levels reported for other plants, usually being lower than 0.5 (32–34). However, almost all rice grain samples (97%) in our study have BAFs for MeHg exceeding 1 (Figure 1). The average BAF of MeHg in rice grain in our data set (5.5–5.6) was higher than reported for other plants (0.55–2.7) (32). Plants with BAF  $\geq 1$  are usually classified as intensive bioaccumulators (32).

A positive correlation between IHg in rice grain and IHg in soil was observed ( $R^2 = 0.19$ ,  $P < 0.01$ , log-transformed) (Figure 2a), suggesting that IHg in paddy soils could serve as a source of IHg in rice. However, the much lower BAFs of IHg compared to those of MeHg (Figure 1) suggest that rice plants may have a similar barrier on the root surface, which blocks IHg uptake through the root system, as other plants (33, 35, 36). Besides, the bioavailability of IHg in soil has been observed generally much lower than that of MeHg (37–39).

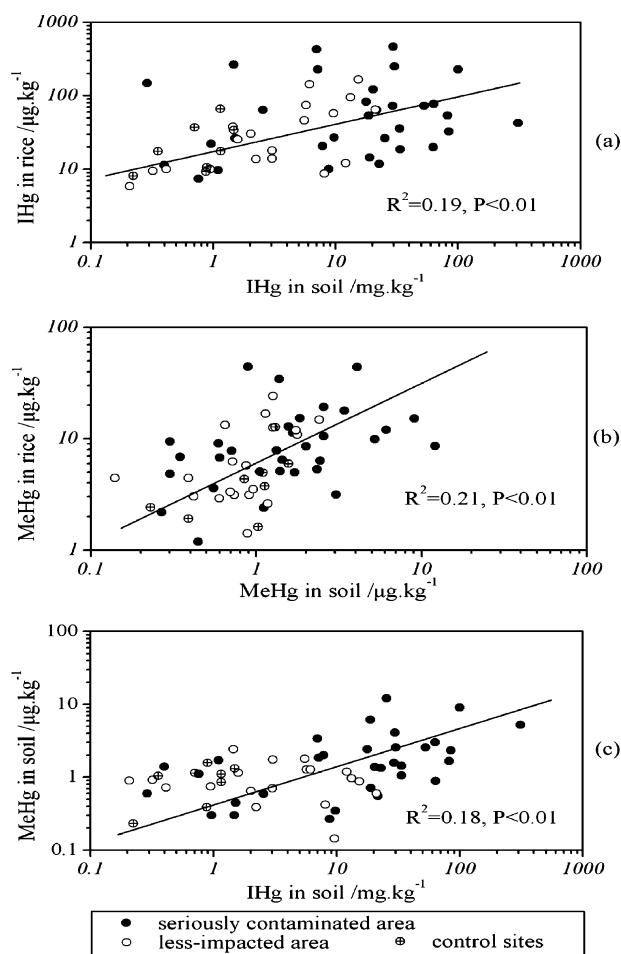
The variation of BAF for Hg may be related to several factors, for example, soil type, pH, organic content, redox potential, and dissolved metal content (33, 40). Rice properties may also be of importance, as genotypic variation, for instance, may affect rice grain accumulation of Hg (41).

A significant positive correlation between MeHg in rice grain and MeHg in soil was observed ( $R^2 = 0.21$ ,  $P < 0.001$ , log-transformed) (Figure 2b), suggesting that MeHg in paddy soil may be the source of MeHg in the rice grain. It is proven that sulfur reducing bacteria (SRB) actively occur in rice paddy soil (42, 43), which are the main contributors of Hg methyl-



**FIGURE 1. Bioaccumulation factors (BAFs, i.e., rice/soil concentrations ratios, BAFs  $\geq 1$  are classified as intensive bioaccumulators (32)) in rice-soil system in Wanshan Hg mined area.**

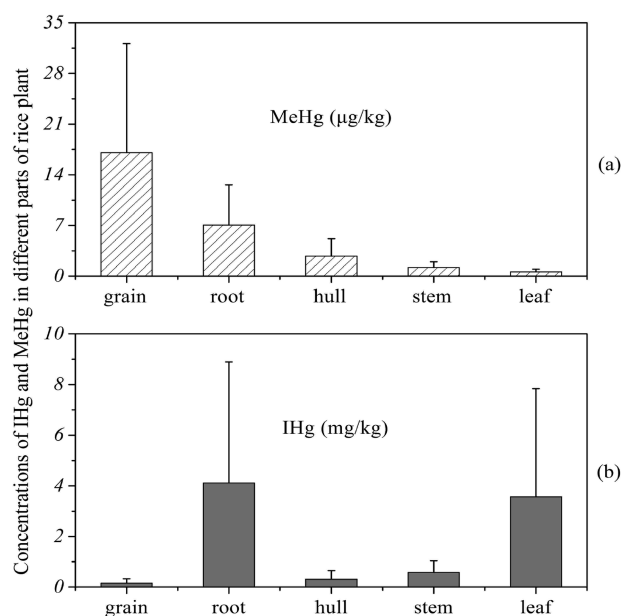




**FIGURE 2.** Relationship between IHg in rice grain and IHg in paddy soil (a), between MeHg in rice grain and MeHg in paddy soil (b), and between MeHg and IHg in paddy soil (c).

lation in the environment (3). In the rice paddy soil samples, MeHg concentrations were significant ( $R^2 = 0.18$ ,  $P < 0.05$ , log-transformed) (Figure 2c) positively correlated to the IHg concentrations, further confirming that Hg methylation processes occurred in rice paddy soil. Besides, Krupp and co-workers (44) have recently found that phytochelatin, small peptides that detoxify heavy metals in rice plants, can sequester  $Hg^{2+}$ , but not MeHg. Hence, different uptake and transport mechanisms for MeHg and IHg should be expected, and may explain the difference in BAFs of IHg and MeHg in the rice plants. Previous studies on other plants have also found that MeHg can be translocated more easily than IHg (32, 45, 46). Our observations clearly show that MeHg in the paddy soil could be more easily taken up and transferred to rice grain compared to IHg.

Organic matter (OM) and pH have been shown to be important factors for MeHg production in aquatic systems in boreal and temperate regions (3). In the present study, however, correlations between soil pH or OM and soil MeHg levels were weak ( $P > 0.05$  for all combinations) (SI Figure S3–S4). This may imply that OM and pH were not key factors explaining the variations in MeHg concentrations in the soil. However, OM values in the soil were positively correlated to concentrations of IHg and MeHg in the rice grain ( $R^2 = 0.15$  and  $0.16$ ,  $P < 0.01$ ) (SI Figure S4), suggesting higher OM content in the soil might be favorable for Hg uptake by the rice plants. However, MeHg BAF did not show significant correlations with OM and pH in soil (SI Figure S5), implying that factors that may control Hg methylation processes in soil have no effects on BAF for MeHg. Other factors, such as genotype, soil type, and the properties of phytochelatin in



**FIGURE 3.** Concentrations of MeHg (a) and IHg (b) in different parts of rice plants from selected sites in Wanshan Hg mining area.

rice plants may control BAF for MeHg in rice. More research is urgently needed on this issue.

One of the five valleys sampled (Gouxi) still had several active, artisanal Hg smelters at the time of sampling. The IHg concentrations in rice samples (mean:  $180 \mu g \cdot kg^{-1}$ ) in this valley were significantly ( $P < 0.01$ ) higher than concentrations in the other valleys (mean:  $20\text{--}50 \mu g \cdot kg^{-1}$ ). The corresponding IHg in paddy soil (mean:  $16 mg \cdot kg^{-1}$ ), however, were not significantly ( $P > 0.05$ ) higher than the other valleys (mean:  $14\text{--}36 mg \cdot kg^{-1}$ ) (SI Figure S2). The BAFs were therefore significantly ( $P < 0.05$ ) higher in Gouxi valley for IHg. However, BAFs of MeHg in Gouxi were not significantly different from other valleys. The higher BAF of IHg may suggest that rice might take up Hg directly from the air through leaf surface in addition to uptake from the roots (47, 48). The atmospheric total gaseous Hg (TGM) concentrations in Gouxi valley was significantly ( $P < 0.01$ ) higher than in the other valleys. Mean TGM concentration in Gouxi valley were generally higher than  $200 ng \cdot m^{-3}$ , and paddy fields near artisanal Hg smelting sites (about 1 km away) could reach a mean concentration of TGM more than  $1000 ng \cdot m^{-3}$  (maximum exceeds  $20\,000 ng \cdot m^{-3}$ ). TGM concentrations in the other valleys were generally lower than  $50 ng \cdot m^{-3}$  (SI Table S1). However, since the OM values in Gouxi valley (mean: 5.1%) were considerably higher than those in the other valleys (mean: 3.0%), and a positive relationship between OM in paddy soil and Hg concentrations in rice grain samples was observed (SI Figure S4), the differences might also partly be related to the OM content in paddy soil.

Analyses of the distribution of Hg species in different parts of rice plants from four selected sampling locations (A9, A20, D4, D12; SI Figure S1) shown that the highest concentration of MeHg was in grain ( $17.1 \pm 15.1 \mu g \cdot kg^{-1}$ ), followed by root ( $7.1 \pm 5.1 \mu g \cdot kg^{-1}$ ) > hull ( $2.8 \pm 2.4 \mu g \cdot kg^{-1}$ ) > stem ( $1.2 \pm 0.8 \mu g \cdot kg^{-1}$ ) > leaf ( $0.6 \pm 0.4 \mu g \cdot kg^{-1}$ ) (Figure 3a). This suggests that rice grain may preferentially accumulate MeHg compared to other parts of the plant. Contrary to MeHg, grain contained the lowest IHg concentrations ( $0.15 \pm 0.18 mg \cdot kg^{-1}$ ), followed by hull ( $0.31 \pm 0.35 mg \cdot kg^{-1}$ ) < stem ( $0.58 \pm 0.46 mg \cdot kg^{-1}$ ) < leaf ( $3.6 \pm 4.3 mg \cdot kg^{-1}$ ) < root ( $4.1 \pm 4.8 mg \cdot kg^{-1}$ ) (Figure 3b). These results support that IHg in the rice plant originated from ambient air through leaf surface absorption in addition to from the soil through root uptake;

while MeHg in rice probably mainly originated from soil through root uptake.

Concentration of MeHg was lower in the leaf than in all other parts of the plant (Figure 3) and the contribution from MeHg in air (even in Hg mined areas) was considered to be negligible (32). Rice paddy soil have been found to be suitable environments for sulfur reducing bacteria (SRB) (42, 43) and hence favorable for Hg-methylation. We suggest that MeHg in the soil was the main source of MeHg in rice, based on the following facts observed in our present study: (1) the distribution trends of MeHg (i.e., grain > root > hull > stem > leaf; Figure 3a); (2) the inconsistent trends of BAF between MeHg and IHg (Figure 1); (3) the positive correlation observed between MeHg in rice grain and in paddy soil (Figure 2b).

Multiple regression analyses showed that MeHg and OM in paddy soil were statistically significant factors ( $P < 0.001$  for both) for MeHg accumulated in the grain ( $\text{MeHg}_{\text{grain}} = 1.35 \text{ MeHg}_{\text{soil}} + 2.0 \text{ OM}_{\text{soil}} - 2.5$ ). The low adjusted  $R^2$  (0.37) indicate the importance of other factors as well (e.g., genotype, soil type, the properties of phytochelatin), that has not been addressed in the present study. More research is needed to scrutinize MeHg distributions in rice plant and to further evaluate potential health impacts on a global scale.

**Risk Considerations.** Residents in Wanshan rarely eat fish ( $1.2 \text{ g} \cdot \text{day}^{-1}$  per capita) (49), and local fish contained low MeHg concentrations (with mean of  $0.060 \pm 0.026 \text{ mg} \cdot \text{kg}^{-1}$  (50)) compared with Chinese national standard limit ( $0.5 \text{ mg} \cdot \text{kg}^{-1}$  (27)). Therefore, Hg exposure through fish consumption in Wanshan does not appear to be of particular concern. A previous pilot study in Wanshan indicated that rice consumption was the major pathway (94–98%) for MeHg exposure to the local inhabitants (13). Based on the concentration data presented here, and statistical dietary data for the region, the calculated MeHg exposure via rice alone was  $0.092 \pm 0.012$  (range: 0.012–0.44)  $\mu\text{g} \cdot \text{kg}^{-1} \text{bw} \cdot \text{day}^{-1}$ . Approximately 34% of the inhabitants in the selected sites exceeded the U.S. EPA reference dose (RfD) of  $0.1 \mu\text{g} \cdot \text{kg}^{-1} \text{bw} \cdot \text{day}^{-1}$ , suggesting some health risk to the local population.

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## Supporting Information Available

Additional text, figures, and tables. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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