

Human Body Burden and Dietary Methylmercury Intake: The Relationship in a Rice-Consuming Population

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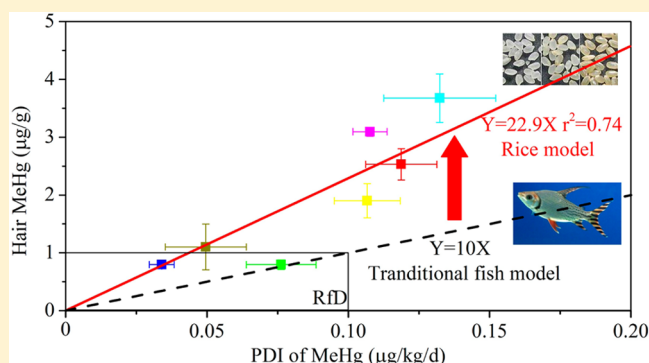
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Supporting Information

ABSTRACT: Rice can be the main route of methylmercury (MeHg) exposure for rice-consuming populations living in area where mercury (Hg) is mined. However, the current risk assessment paradigm for MeHg exposure is based on epidemiological data collected from fish-consuming populations. This study was designed to evaluate the relationship between dietary MeHg intake and human body burden in a rice-consuming population from the Wanshan Hg mining area in China. Hair MeHg concentrations averaged $2.07 \pm 1.79 \mu\text{g/g}$, and the average blood MeHg concentration across the study area ranged from 2.20 to $9.36 \mu\text{g/L}$. MeHg constituted $52.8 \pm 17.5\%$ and $71.7 \pm 18.2\%$ of total Hg (THg) on average in blood and hair samples, respectively. Blood and hair MeHg concentrations, rather than THg, can be used as a proxy of human MeHg exposure. Hair MeHg levels showed no significant monthly variation; however, hair THg can be impacted by inorganic Hg exposure. The toxicokinetic model of MeHg exposure based on fish consumption underestimated the human hair MeHg levels, and this may be a consequence of the high hair-to-blood MeHg ratio (361 ± 105) in the studied rice-consuming population. The use of risk assessment models based on fish consumption may not be appropriate for inland mining areas where rice is the staple food.



INTRODUCTION

The toxicity of methylmercury (MeHg) has been extensively documented by the World Health Organization,¹ the United States Environmental Protection Agency (USEPA),² the Agency for Toxic Substances and Disease Registry,³ and the United States National Research Council⁴ and has been reviewed by Clarkson,⁵ Clarkson and Magos,⁶ Mergler et al.,⁷ and Driscoll et al.⁸ Exposure of the general population to MeHg occurs primarily through the consumption of fish and marine mammals.⁷ MeHg contamination in fish poses a particular challenge to public health because of the documented nutritional benefits from fish for human health.⁷ MeHg in fish tissue has been identified as being bound to the thiol group of cysteine in fish protein,⁹ which is not removed or destroyed by the cooking or cleaning processes. The maximum level of MeHg in fish recommended by the Joint FAO/WHO Food Standards Programme CODEX Committee on Contaminants in Foods¹⁰ is $1.0 \mu\text{g/g}$ for predatory fish and $0.5 \mu\text{g/g}$ for other fishery products. This guideline has been adopted by most countries.

The physiologically based pharmacokinetic (PBPK) model¹¹ and the one-compartment model^{12,13} have been used to link intake dose and blood concentration. Even though these

relationships are well-established, significant variability can be found in the reported parameters.¹⁴ A concentration ratio of 250:1 for mercury (Hg) in hair to mercury in blood is frequently reported,^{1,2} but uncertainty also existed in this value, with a range of 140 to 370.^{1,15} Canuel et al.¹⁶ reported big differences between hair Hg concentration and the expected value based on the calculated daily oral intake. Wang et al.¹⁷ and Goodrich et al.¹⁸ found an effect of genetic polymorphism on Hg concentrations in biomarkers, even at low levels of Hg exposure.

On the basis of the studies conducted in the Faroes Islands (cord blood Hg concentration of $58 \mu\text{g/L}$), USEPA set the limit of $0.1 \mu\text{g/kg/day}$ as the reference dose (RfD) for MeHg.² According to the calculations made in the Faroe Islands study and in another from the Seychelles ($12 \mu\text{g/g}$ Hg concentration in maternal hair), the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a provisional tolerable weekly intake (PTWI) for MeHg at $1.6 \mu\text{g/kg/week}$.¹⁹ The quantification

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of these intake values is important because the effect of low-level MeHg exposure on neurological development has been quantified. A loss of 0.18 intelligence quotient (IQ) points is thought to be associated for each part per million increase of maternal hair Hg.^{20,21}

Recent studies have highlighted the fact that rice can accumulate high levels of MeHg in mercury mining areas in Guizhou, Southwestern China,^{22–24} and the dynamic mechanism of MeHg translocation and accumulation in rice plant has been investigated.^{25,26} Rice, rather than fish, is the main route of human MeHg exposure in the Wanshan Hg mining area,²⁷ Guizhou Province,²⁸ and inland area of southern China.²⁹ Estimations of the probable daily intake (PDI) of MeHg from rice consumption indicate that rice consumption is a potential human health risk in Hg mining areas.^{27,28,30}

Rice lacks many of the beneficial micronutrients associated with fish (e.g., n-3 long-chain polyunsaturated fatty acid (n-3 LCPUFA), selenium (Se), and essential amino acids),³¹ and, therefore, the current RfD and PTWI from fish consumption may underestimate the health effects of MeHg exposure from rice consumption. Balancing the risks and benefits of fish consumption has become an increasingly important goal of fish consumption advisories. Ginsberg and Toal³² provided an integrated analysis for MeHg and n-3 LCPUFA that evaluated the net effect of fish consumption on a species-by-species basis. Zhang et al.³³ proposed a new criterion for assessing MeHg exposure and Se intake based on Se–Hg interactions and the physiology and toxicology of Se. In general, the interaction between MeHg and micronutrients is unknown. Such interactions may impact the dose–response relationship between the MeHg intake and the body burden in a rice-consuming population.

This study was designed to evaluate the relationship between MeHg intake and blood and hair MeHg levels in a rice-consuming population at the Wanshan Hg mining area. It also aimed to develop a risk assessment paradigm for MeHg exposure among rice consumption populations. In a companion paper, inorganic Hg (IHg) exposure, renal effects, and possible pathway were evaluated in the same study population.³⁴

MATERIALS AND METHODS

Study Area. The Wanshan Hg mining area, located in the eastern part of Guizhou province, was the largest Hg mine in China. Large scale Hg mining activities operated in the area for over 50 years, leading to serious Hg contamination to the local environment.^{35–40} The Hg adits and historical retorts are situated upstream of four rivers in the region (Figure S1 in the Supporting Information), and significant quantities of mine wastes are the primary Hg contamination source to rivers.^{36,37,41} Rice is widely cultured throughout the valleys in this region, and the river water is the main source for irrigation.

Wanshan County consists of five towns (namely Wanshan, Huangdao, Xiayi, Aozhai, and Gaolouping) and covers an area of approximately 338 km² (Figure S1 in the Supporting Information). The population in Wanshan County in 2012 was 68 000, and the rural population constituted about 80% of the total according to the local government's web site. The local economy is undeveloped, and the per capita gross domestic product was 14 914 RMB (2400 USD) in 2011,⁴² which was about half of the national average in China. A total of seven sites in Xiayi and Aozhai were selected for survey in the current study and were distributed along the Aozhai and Xiaxia River with different gradients from the Hg pollution source (Figure S1 in the Supporting Information).

Sample Collection. Sampling was conducted in December of 2012. Participants were recruited with the criteria that they had been a local resident for over 3 months. The recruitment strategy in the survey was to recruit 10–20% of whole population at each site. All of the participants participated voluntarily. The recruitment period lasted 2 days. A questionnaire was used to obtain the basic information on age, body weight, profession, history of involvement in artisanal Hg mining activity, dental fillings, smoking and alcohol drinking habits, illness, and the amount of daily rice consumption. Daily rice intake (g/day) was quantified by participants according to their eating habits and the whole-family monthly or annual rice consumption figures reported by the housewives.

Residents of the Wanshan mining area seldom eat fish, and consumption is similar to the average daily fish intake of 1.2 g/day for Guizhou rural residents.⁴² A previous study has shown that the MeHg concentrations in fish samples collected from Wanshan Hg mine were relatively low, with a mean of 0.060 µg/g.⁴³ Fish samples were not collected in the current study because these figures indicate that fish is not a significant dietary source of MeHg in the population in Wanshan.^{28,29,44}

Hair samples were cut with stainless steel scissors from the occipital region of the scalp of each participant, bundled together in polyethylene bags, and then transferred to the laboratory. A total of 11 female subjects with long hair were selected from across the study area for sequential Hg analysis. Hair strands were cut into sequential 12 cm sections from the scalp to examine the monthly variations of hair Hg levels in the past year (the growth rate of hair was estimated to be 1 cm per month⁴). Venous blood samples (5 mL) were collected from each participant in prepared EDTA vacuum tubes. Blood samples were stored on ice during the fieldwork and transferred to –20 °C upon receipt at the laboratory. Raw rice samples were collected from each participating household.

The present study obtained ethics approval from the Institute of Geochemistry, Chinese Academy of Sciences. All participants signed an informed consent form before engaging in the survey.

Analytical Methods. The portion of hair within 3 cm from the scalp was selected for total Hg (THg) and MeHg analysis. Hair samples were washed with nonionic detergent, distilled water, and acetone according to Li et al.⁴⁵ and dried in an oven at 60 °C overnight. Rice samples were air-dried, crushed, and sieved to 150 meshes. The THg concentrations in the hair and rice samples were analyzed by the RA-915+ Hg analyzer coupled with the PYRO-915+ attachment (Lumex) using thermal decomposition and Zeeman atomic absorption spectrometry. Whole blood samples (0.1 mL) were digested with a mixture of nitric and sulfuric acid (v/v 4:1) at 95 °C for 3 h. The digests were analyzed for THg concentration following the sequence of BrCl oxidation, SnCl₂ reduction, purging, gold trapping, and cold vapor atomic fluorescence spectrometry (CVAFS).⁴⁶

Hair, rice, and blood samples were digested using the KOH–methanol/solvent extraction technique.⁴⁷ MeHg concentrations were measured using the aqueous ethylation, purging, trapping, and GC–CVAFS detection (Brooks Rand MERX) method. Detailed information on the quality control systems adopted in this work is listed in Table S1 in the Supporting Information.

Calculation of PDI. To estimate the daily MeHg intake from rice consumption, we calculated the PDI values for each participant at the seven sites using eq 1:

$$\text{PDI} = (C \times \text{IR} \times 10^{-3})/\text{bw} \quad (1)$$

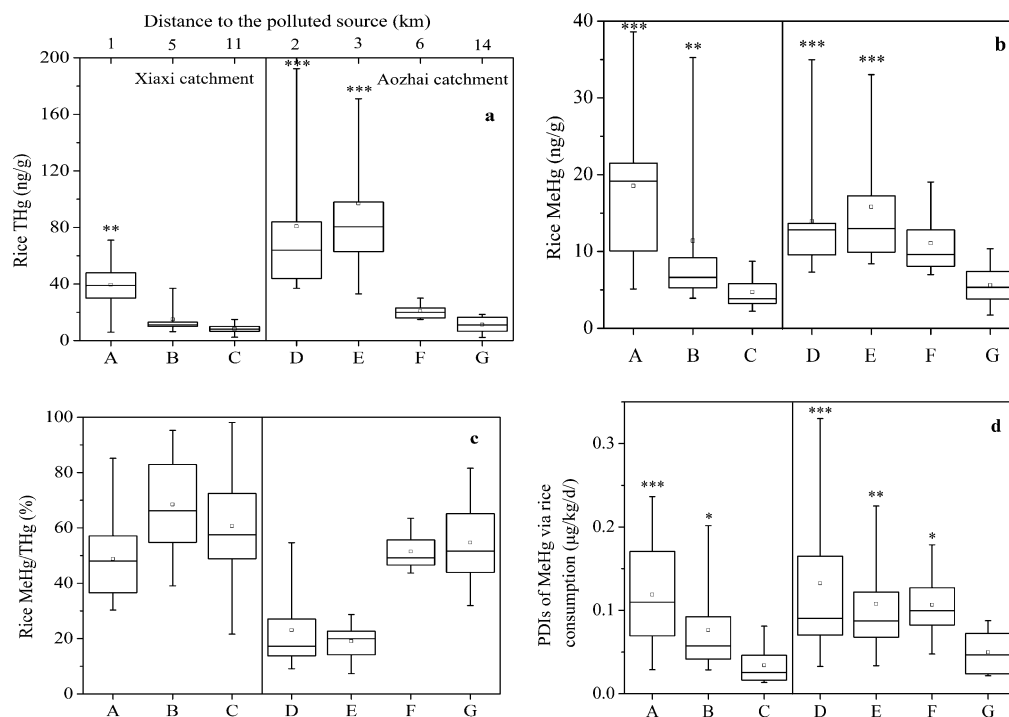


Figure 1. Comparison of rice Hg concentrations and PDIs of MeHg via rice consumption at different sites. ***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$. Data from sites A and B were compared with data from site C; data from sites D–F were compared with data from site G. Each box represents the interquartile range (25th and 75th percentile); the band near the middle of the box is the 50th percentile (the median), the whisker represents the 5th and 95th percentile, and the dot under the box is the mean.

where PDI is given in μg per kg of body weight per day ($\mu\text{g}/\text{kg}/\text{day}$); bw is body weight (kg), C is the MeHg concentration in rice (ng/g), and IR is the daily intake rate of rice (g/day), which was obtained from the questionnaire of each participant.

Data Analysis. All data were analyzed using SPSS 19.0 for Windows. The characteristics of Hg concentration were described by the mean \pm standard deviation (SD) and examined using descriptive statistics. Mean Hg concentrations in rice, hair, and blood collected from different sites were compared using ANOVA. The relationships between rice, hair, and blood Hg concentrations and the PDI of MeHg were analyzed using the Pearson correlation analysis. The results of the statistical tests were considered as statistically significant when $p < 0.05$.

RESULTS

Basic Information. Descriptive statistics of the study population and information on rice consumption are shown in Table S2 in the Supporting Information. The recruited participant numbers in the survey accounted for 10–20% of the population at each site except sites C and F, which were big towns with large populations. In total, 110 households were contacted in this survey, and about one-third (58/168) of the participants were from the same households. Most village residents cultivated rice and were self-sufficient in this food, while the residents in the towns purchased rice from the market (for instance, Site C). The average rice ingestion rate was 428 ± 157 g/day, which is comparable with that for Guizhou's rural population (371 g/day) as recorded by the official government statistical yearbook for 2012.⁴²

Rice Hg and PDIs of MeHg. The concentrations of THg and MeHg in the rice samples collected from the seven sites are shown in Figure 1. The average THg concentrations in all rice samples was 42.4 ± 52.7 ng/g, and 53% of samples exceeded the

national limit (20 ng/g).⁴⁸ The MeHg concentrations in all rice samples averaged 11.7 ± 8.84 ng/g, and MeHg constituted $44.8 \pm 23.9\%$ of THg. The average percentages of THg as MeHg in rice samples from sites D and E were relatively low ($23.0 \pm 16.7\%$ and $19.1 \pm 8.44\%$, respectively), as shown in Figure 1c. A significant correlation between THg and MeHg concentrations in all rice samples was found ($r = 0.44$, $p < 0.001$).

The PDIs of MeHg through rice consumption for the residents at the seven sites are presented in Figure 1d. The average PDI of MeHg from rice consumption ranged from 0.050 to $0.132 \mu\text{g}/\text{kg}/\text{day}$.

Hair and Blood Hg. The distribution of THg and MeHg concentration in hair samples from the seven sites is shown in Figure 2. The average THg concentration in hair ranged from 1.33 to $5.07 \mu\text{g}/\text{g}$ (Figure 2a); for MeHg, it ranged from 0.79 to $3.67 \mu\text{g}/\text{g}$ (Figure 2b). MeHg constituted $71.7 \pm 18.2\%$ of THg in all hair samples on average and was the main form of Hg in hair. A significant correlation ($r = 0.84$, $p < 0.001$) between hair THg and MeHg concentrations was observed.

In the total population, the overall average blood THg concentration was $12.2 \pm 15.0 \mu\text{g}/\text{L}$ (2.15–30.8, 95% CI), as shown in Figure 2c. The average blood MeHg concentration at different sites ranged from 2.20 to $9.36 \mu\text{g}/\text{L}$ (Figure 2d). MeHg constituted $52.8 \pm 17.5\%$ of THg in all blood samples on average.

In this study, a significant positive correlation ($r^2 = 0.83$, $p < 0.001$) was observed between the human blood MeHg concentration and the PDI of MeHg via rice consumption (Figure 3a). A significant positive correlation ($r^2 = 0.74$, $p < 0.001$) was also observed between human hair MeHg concentration and the PDI of MeHg via rice consumption (Figure 3b).

Monthly Variation of Hair Hg. The monthly variation of hair THg concentration in the 11 subjects selected for sequential analysis is shown in Figure 4a. Except for D3 and E13, the hair

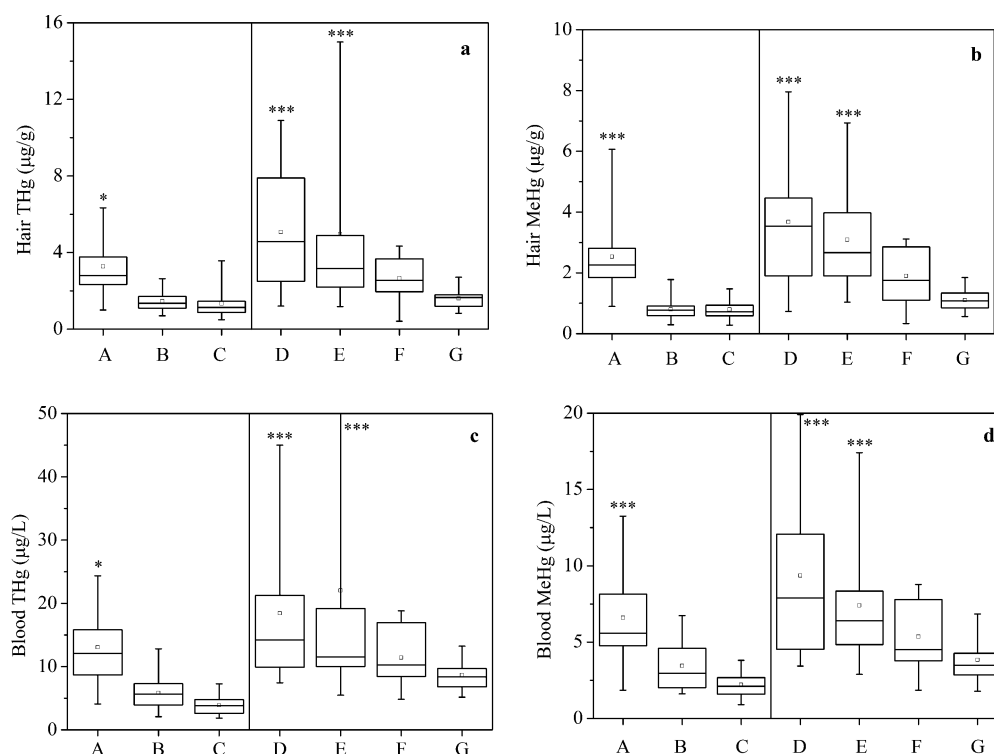


Figure 2. Comparison of hair and blood Hg in the population at different sites. ***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$. Data from sites A and B were compared with data from site C; data from sites D–F were compared with data from site G.

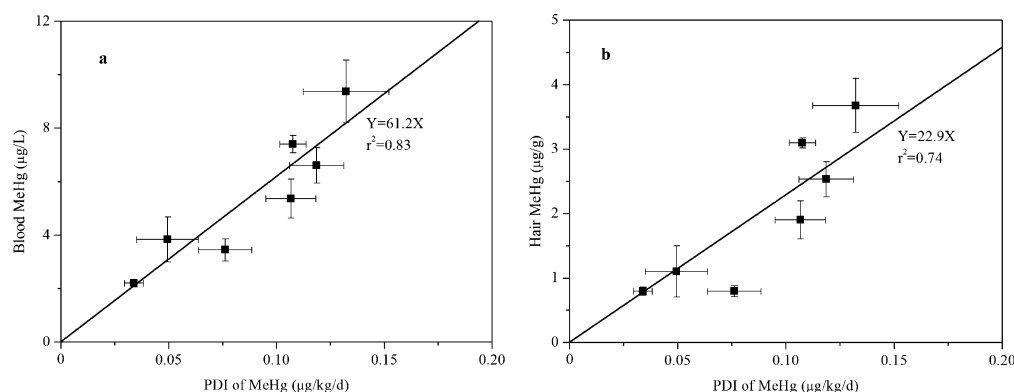


Figure 3. Relationship between human blood and hair MeHg and the PDIs of MeHg via rice consumption at different sites.

THg concentration was stable with time (Figure 4a). The coefficients of variation (CV) of hair THg concentration at different months were less than 10%. The percentage of THg as MeHg in these nine hair samples averaged 88.8%, confirming that MeHg was the main form of THg in the hair samples. In two hair samples (D3 and E13), the hair THg concentration varied between months, but the hair MeHg concentration each month was stable ($CV < 15\%$; Figure 4b). This indicates that participants D3 and E13 were exposed to IHg. An analysis of urine for these two subjects showed highly elevated Hg (17.1 and 33.7 µg/g creatinine, respectively), confirming the exposure to IHg.³⁴ Our data indicate that the annual levels of human MeHg exposure in Wanshan are stable, which is likely due to the stability of the MeHg concentration in the consumed rice across the year.

DISCUSSION

Biomarker of MeHg Exposure. Hair and blood THg concentrations are both considered good biomarkers of human

MeHg exposure in the population that consumes fish.⁷ The mean half-life of MeHg in human blood is about 50–70 days, and blood reflects relatively short-term exposures to Hg.⁴ Hair reflects the average Hg exposure of an individual over the growth period of the hair segment.⁷ For the fish-eating population, MeHg is the main form of Hg in the hair and blood and will constitute between 80% and 98% of THg.¹ Therefore, THg concentrations in blood and hair samples are widely used as a proxy of MeHg concentrations in human MeHg exposure models.

The percentages of THg as MeHg in human hair and blood samples in this study were much lower than the above range (80–98%), with an average of $71.7 \pm 18.2\%$ and $52.8 \pm 17.5\%$, respectively. The ratios varied significantly between individuals (Figure S2 in the Supporting Information). However, hair and blood MeHg concentrations are justified as biomarkers of human MeHg exposure in this study. Our results indicate that local residents are coexposed to IHg and MeHg. Elevated IHg exposure is associated with the consumption of IHg-contaminated rice and

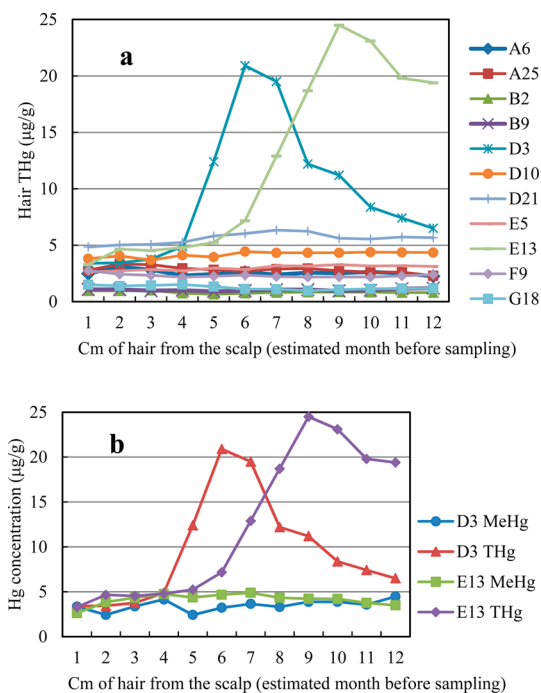


Figure 4. Monthly variation of hair Hg concentrations in the selected subjects.

vegetables.³⁴ In the context of coexposure to IHg and MeHg, IHg analysis will overestimate human MeHg exposure if adopting blood and hair THg concentrations.

Geographical Difference. The concentration of THg and MeHg concentration in rice samples reduced within the two catchments as a function of the distance from the pollution source (Figure 1). The concentration of both THg and MeHg in rice samples was elevated in the upstream region (sites A, D, and E) seriously impacted by the mine wastes relative to the downstream sampling points.^{36,37} Hg in the paddy soil could serve as a source of Hg in rice both for IHg and MeHg.²⁴ The average values for PDI at each site showed a similar trend of reduction with increasing distance from the point-source of Hg contamination within the two catchments (Figure 1d).

Almost 73% (122/168) of blood THg concentrations exceeded 5.8 µg/L, which is the blood Hg level equivalent to the current RfD set by USEPA. Individuals from sites close to the mine waste heaps (A, D, and E) showed significantly higher hair and blood THg and MeHg levels than those at site C and G, who recorded values in agreement with the regional background (Figure 2c).

Hair-to-Blood Ratio. A “normal” hair-to-blood Hg concentration ratio of 250:1 is frequently cited in the literature;² however, uncertainty over the accuracy of this ratio exists in

many reports. In one study, the median ratios in Faroese children were 370 at 7 years of age and 264 at 14 years.⁴⁹ The median ratio in a Swedish population was 373,⁵⁰ and the mean ratio after an adjustment of time lag was 344 ± 54 (with a range of 263–478) in Japan.⁵¹ In the risk assessment for MeHg, both the National Academy of Science National Research Council (NAS–NRC) and JECFA adopted an uncertainty factor of 3.2 on the toxicokinetic variability.^{4,19}

The mean ratio of hair-to-blood MeHg concentration in our study (expressed as µg MeHg/g hair to mg MeHg/L blood) was 361 ± 105, and the 5th and 95th percentiles were 178 and 538, respectively (Figure S3 in the Supporting Information). The ratios of hair-to-blood THg concentrations were calculated as well for comparison (Figure S3 in the Supporting Information); the mean ratio was 268 ± 112, and the 5th and 95th percentiles were 127 and 493, respectively (Figure S3 in the Supporting Information). The percentage of THg as MeHg in hair was much higher than that in blood (average of 72% to 52%), which resulted in an elevation of hair-to-blood ratios for MeHg relative to THg. Significant correlations were found between the hair and blood MeHg and THg concentration in the study population (Figure S4a for MeHg and Figure S4b for THg in the Supporting Information). The results from this study (361 ± 105) were 44% higher than the average value of 250, which indicates that the reported average or normal value may underestimate the MeHg distribution between human hair and blood in the study population.

Relationship between PDI and Body Burden. The one-compartment model calculated the steady-state Hg concentration in blood (*C*) in µg/L from the average daily dietary intake (in µg Hg per kg of body weight per day) as shown in eq 2:^{1,2}

$$C = \frac{d \times A \times f \times bw}{b \times V} \tag{2}$$

where *d* represents the PDI of MeHg in µg/kg/day, *A* is the absorption factor (which is assumed to be 0.95),² *f* is the absorbed dose found in blood (value of 0.05),² *bw* is the body weight in kg, *b* is the elimination constant (0.014/day),² and *V* is the blood volume (L, 8% of bw).⁵²

Using eq 2, modeled MeHg concentrations in blood can be obtained from the PDI values. Considering the hair-to-blood ratio as the average of 250:1, modeled hair MeHg concentrations (*H*) also can be obtained from the PDI values for individuals.

Table 1 presents the calculated MeHg PDIs at the seven sites and the comparison of the simulated value with the measured blood and hair MeHg concentrations. There was good agreement between the measured blood and hair MeHg levels and the estimated values at site B. However, the modeled blood and hair MeHg levels for other sites were significantly lower than the measured concentrations.

Table 1. Simulation Runs for Blood and Hair MeHg at the Seven Sites of This Study

site	PDI of MeHg (µg/kg/day)	blood MeHg (µg/L)			hair MeHg (µg/g)		
		modeled	measured	variability (%)	modeled	measured	variability (%)
A	0.119 ± 0.068	5.03	6.60	76	1.26	2.53	50
B	0.076 ± 0.051	3.23	3.44	94	0.81	0.79	103
C	0.034 ± 0.025	1.44	2.20	65	0.36	0.80	45
D	0.132 ± 0.105	5.61	9.36	60	1.40	3.67	38
E	0.108 ± 0.076	4.57	7.40	62	1.14	3.09	37
F	0.107 ± 0.037	4.52	5.36	84	1.13	1.90	59
G	0.050 ± 0.030	2.10	3.83	55	0.53	1.10	48

A significant positive correlation was obtained between human hair MeHg concentrations (H) and PDIs of MeHg via rice consumption (d) at the different sites (Figure 3b) with the regression equation quantified as $H = 22.9d$. The corresponding reported model for a fish-eating population is $H = 10d$.² In practical terms, this means that an RfD of 0.1 $\mu\text{g}/\text{kg}/\text{day}$ corresponds to a hair MeHg concentration of 1 $\mu\text{g}/\text{g}$. The regression coefficient for the model derived from our study was 2.3 times greater than that for the fish model, indicating that hair MeHg levels are highly elevated at the same exposure dose for the population consuming MeHg-contaminated rice relative to fish. The MeHg toxicokinetics model based on fish consumption is therefore not suitable for risk assessment of MeHg exposure via rice consumption.

The intrapopulation variability of human metabolism of MeHg has been reported in many epidemiologic studies. In the risk assessment of MeHg exposure, NAS–NRC set an uncertainty factor of 10 to account for variability and uncertainty in toxicokinetics and toxicodynamics, and JECFA used an overall uncertainty factor of 6.4 to address variability in both toxicokinetics and toxicodynamics. The pharmacokinetic constants used in the one-compartment model also vary extensively between different studies.¹² For instance, the whole-body half-life of MeHg is generally considered as 70 days (with the corresponding elimination constant of 0.014/day), but this number is reported to vary between 44 and 80 days¹² or roughly +15%/–63%. The mean fraction of absorbed dose in blood (f) also ranges from 5% to 7.7%.^{1,2} The high ratio of hair-to-blood MeHg in the study population also contributes to uncertainties, and the average of 361 was 44% higher than the general average of 250. Canuel et al.¹⁶ reported big differences between hair Hg concentration and the expected value based on the calculated daily oral intake. Wang et al.¹⁷ and Goodrich et al.¹⁸ found an effect of genetic polymorphism on Hg biomarker concentrations even at low levels of Hg exposure.

Compared with fish, rice is of poor nutritional quality and lacks specific micronutrients identified as having health benefits (e.g., n-3 LCPUFA, Se, and essential amino acids).³¹ A wide variety of foods and nutrients may impact the absorption, distribution, and elimination of MeHg in human body⁵³ and the inconsistency of the MeHg toxicity observed in different populations is commonly attributed to the possible effects of dietary modulation.⁵³ Our results indicate that the RfD and PTWI based on fish consumption may underestimate the risks caused by MeHg exposure from rice consumption.

Implication for Public Health. The hair MeHg concentrations obtained in this study ($2.07 \pm 1.79 \mu\text{g}/\text{g}$) were comparable with those previously reported for residents at the Wanshan Hg mine ($1.85 \pm 1.16 \mu\text{g}/\text{g}$)²⁷ and Wuchuan Hg mine ($1.25 \pm 0.74 \mu\text{g}/\text{g}$)⁵⁴ but were lower than those for residents in the Tongren Hg mines (5.24 ± 2.80 and $3.76 \pm 1.73 \mu\text{g}/\text{g}$ for workers and residents, respectively)³⁰ and for fishers in Zhoushan Island ($3.8 \mu\text{g}/\text{g}$).⁵⁵ The blood THg concentrations in this study ($12.2 \pm 15.0 \mu\text{g}/\text{L}$) were comparable with the mean of blood THg in local residents from the Chatian Hg mine ($6.09 \pm 3.26 \mu\text{g}/\text{L}$)⁵⁶ and lower than the serum Hg in residents also from the Wanshan Hg mine (with a mean of $38.5 \mu\text{g}/\text{L}$).⁵⁷

In this study, hair MeHg levels are highly elevated at the same exposure dose for the population consuming Hg-contaminated rice relative to fish. We propose that hair MeHg concentrations should be used as a more accurate measure of human exposure to Hg in the study population. The equivalent hair MeHg levels of 1.0 and 2.3 $\mu\text{g}/\text{g}$ recommended by USEPA and JECFA have been

adopted for the risk assessment of MeHg exposure on a developing fetus. The total population in the Wanshan area is 68 000. For women of child-bearing age (15–44 years), the distribution of hair MeHg concentration within the population was calculated as $>1 \mu\text{g}/\text{g}$ for 5600 inhabitants (8.2% of total population) and $>2.3 \mu\text{g}/\text{g}$ for 1400 inhabitants (2.1% of total population). In addition to Wanshan, there are 11 other Hg mining and smelting areas in Guizhou,⁵⁸ with populations totaling approximately 320 000. Considering women of child-bearing age (15–44 years), there are 26 240 inhabitants with hair MeHg concentrations higher than 1 $\mu\text{g}/\text{g}$ and 6720 inhabitants with hair MeHg concentrations higher than 2.3 $\mu\text{g}/\text{g}$.

Rice is the staple food of more than half the world's population.⁵⁹ MeHg accumulation in rice and the related health risks should be a focus of concern in Hg-polluted areas (e.g., Hg mines, abandoned chloralkali facilities, and artisanal and small-scale gold mines) where rice is a staple food. The findings obtained in this study can be used to advise human MeHg exposure and risk assessment through rice consumption in these areas.

■ ASSOCIATED CONTENT

● Supporting Information

Details of QA and QC protocol. Tables showing the certified reference materials and descriptive statistics of the study population and rice consumption information. Figures showing the location of sampling sites, a comparison of Hg percentages, hair-to-blood ratios of THg and MeHg concentrations, and the relationship between hair and blood MeHg/THg. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b00195.

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Notes

The authors declare no competing financial interest.

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