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Human co-exposure to mercury vapor and methylmercury in artisanal mercury mining areas, Guizhou, China

Ping Li [a], Xinbin Feng [a,*], Lihai Shang [a], Guangle Qiu [a], Bo Meng [a,b], Hua Zhang [a,b], Yanna Guo [a], Peng Liang [a]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[b] Graduate University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Mercury (Hg) concentrations were determined in human urine and hair samples from Gouxi (GX, $n=25$) and Laowuchang (LWC, $n=18$), Tongren, Guizhou, China, to evaluate human exposure from artisanal Hg mining. Geometric means of urinary Hg (U-Hg) were 216 and 560 $\mu g\,g^{-1}$ Creatinine ($\mu g\,g^{-1}$ Cr) for artisanal mining workers from GX and LWC, respectively, and clinical symptoms (finger tremor) were observed in three workers. The means of hair Me-Hg concentrations were 4.26 $\mu g\,g^{-1}$ (1.87–10.6 $\mu g\,g^{-1}$) and 4.55 $\mu g\,g^{-1}$ (2.29–9.55 $\mu g\,g^{-1}$) for the population in GX and LWC, respectively. Significant relationship was found between estimated rice Me-Hg intake and hair Me-Hg levels ($r=0.73$, $p<0.001$). Co-exposure to Hg vapor and Me-Hg may pose health risks for the study population.

1. Introduction

Mercury (Hg) is a highly toxic element and can cause significant adverse effects on human health, but the toxicity depends on its chemical form.

For Hg vapor exposure, dental amalgam filling is the predominant source in the general population and most of research concludes that there is no health risk associated with this exposure (Clarkson, 2002). But for occupational exposure, such as workers in dental clinics, chloralkali plants, fluorescent lamp factories, Hg mines and artisanal and small-scale gold and silver mining operations, inhalation of Hg vapor may cause a variety of adverse effects. The most classic symptoms are found in the central nervous system (tremor, and mental changes) and the kidneys (e.g., proteinuria) (WHO, 1991). Urine Hg (U-Hg) measurements are widely used for assessment of inorganic Hg (mainly Hg vapor) exposure in humans, because it closely reflects Hg levels in the kidney (Clarkson et al., 1988; Barregard, 1993).

Human exposure to Me-Hg is mainly related to fish consumption (WHO, 1990; USEPA, 1997). It poses a particular challenge to public health, because fish is a highly nutritious food, and also constitutes an important global commodity (Mergler et al., 2007). The toxicity of Me-Hg is much higher than that of inorganic Hg. Me-Hg exposure and related health effects in humans has gained much of the attention of researchers since it is a worldwide concern (Mergler et al., 2007).

The nervous system is the primary target organ for Me-Hg poisoning, which can result in marked distal sensory disturbances, constriction of visual fields, ataxia, dysarthria, auditory disturbances, and tremor (Harada, 1995; Clarkson et al., 2003). The poisoning in Minamata Bay brought attention to the risk from fetal exposure, since the developing central nervous system is more sensitive to damage from Me-Hg than the adult nervous system. Recently there is some new evidence between Me-Hg exposure and cardiovascular effects (Chan and Egeland, 2004; Stern, 2005), reproductive effects (Sakamoto et al., 2001; Itai et al., 2004), and immune system effects (Hultman and Hansson-Georgiadis, 1999; Haggqvist et al., 2005), but more quantitative dose–response assessment should be undertaken. Hair and blood Hg levels are widely used to monitor of Me-Hg exposure (NRC, 2000).

Hg vapor and Me-Hg are separately potent neurotoxins and differ considerably in their toxico-kinetics and toxico-dynamics. But both of them are effortlessly absorbed and both are high lipophilic substances, which easily cross the blood–brain barrier exposing the developing fetus. Of significant concern, the human fetus appears especially vulnerable to Me-Hg. If the ultimate toxic form of Me-Hg is its inorganic Hg metabolite, it suggests that the dose of inorganic Hg to the brain from elemental Hg exposure and Me-Hg might be cumulative (NRC, 2000). In animal data, co-exposure to Hg vapor and Me-Hg show an additive effect

* Correspondence author. Fax: +86 851 5891609.
E-mail addresses: fengxinbin@vip.skleg.cn, xinbin.feng@mail.gyig.ac.cn (X. Feng).

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on the developing central nervous system (Fredriksson et al., 1996). But there are no data on the combined exposure to Hg vapor and Me-Hg exposure for humans, despite concerns that combined exposure may elevate risk via additive or synergistic mechanisms.

This study was designed to evaluate co-exposure to Hg vapor and Me-Hg in the population in GX and LWC, Tongren, Guizhou, China.

Our previous study confirmed that the artisanal Hg mining in the study area have emitted significant quantities of Hg to the ambient air (Li et al., 2009) resulting in serious Hg contamination to stream waters, soils (Li et al., 2008a), and rice (Qiu et al., 2008). The artisanal mining workers in Wuchuan Hg mining areas were seriously exposed to Hg vapor through inhalation (Li et al., 2008b; Iwata et al., 2007). Previous studies demonstrated that rice grain is an intensive bioaccumulator of Me-Hg, but not of Inorganic Hg (Qiu et al., 2008; Zhang et al., 2010a; Meng et al., 2010). High Me-Hg levels were found in the rice collected from GX (ranging from 22.3 to 174 ng/g with an average of 62.3 ng/g) and LWC (ranging from 10.1 to 43.1 ng/g with an average of 26.3 ng/g) (Qiu et al., 2008). The population in Hg mining area is exposed to Me-Hg through rice consumption and they are at the potential risk of Me-Hg exposure (Feng et al., 2008; Qiu et al., 2008; Li et al., 2008c; Zhang et al., 2010b). Hence the population in the study area may be at the risk of co-exposure to Hg vapor through inhalation and Me-Hg via rice eating.

2. Materials and methods

2.1. Sample collection

The workers involved in artisanal Hg mining and local residents in GX and LWC, Tongren District, were selected for the study. GX and LWC are situated in Tongren District, Eastern Guizhou, which was described in detail in the previous study (Li et al., 2008a). For the economic benefits, local farmers take part in the artisanal Hg mining during slack farming season, although it is illegal in China. The Hg ores for artisanal Hg mining in GX are provided by local small scale Hg mines, while in LWC Hg recovery is from abandoned catalysts, used in polyvinyl chloride (PVC) manufacture.

The sampling campaign was conducted in December 2006. We selected 25 and 18 participants in GX and LWC, respectively, for the investigation. The only criterion to select the participants was that they were the local residents and lived in their homes at least for 3 months before the sampling. The recruitment strategy was to include as many participants as possible. Most of the young people in the villages went out to do labor work and some workers were not willing to participate in the survey because the artisanal mercury mining was illegal in China, and therefore these numbers were the maximum numbers we could get from each site.

Urine and scalp hair samples were collected for investigating Hg exposure levels. The urine samples were collected in pre-cleaned plastic centrifugal tubes, hermetically sealed and kept at 4 °C until analysis. Hair samples were cut with a stainless steel scissors from the occipital region of the scalp, bundled together with scrip, placed, and sealed in polyethylene bags, properly identified and taken to the laboratory for analysis. Samples of rice, which is a food staple for the participants, were collected from the family of each participant for Hg analysis and daily Hg intake was estimated. All rice samples consumed by the local residents were cultivated from their own land.

A questionnaire was utilized to collect information on residential history, occupational history, dietary habit, life style (smoking and alcohol drinking), and health history. The present study obtained the ethics approval from Institute of Geochemistry, Chinese Academy of Sciences. All participants joined the investigation on a voluntary basis.

2.2. Analytical method

Hair samples were washed with ultrasound successively in nonionic detergent, distilled water, and acetone, and dried in an oven at 60 °C overnight. The strict procedure was designed to avoid exogenous Hg contamination in the hair, since the study population was in a high level of Hg vapor exposure living condition. Urine, hair, and rice samples were digested in a water bath (95 °C) with a fresh mixed acid of HNO_3:H_2SO_4 (4:1, v/v) for T-Hg analysis (Horvat et al., 1991). T-Hg concentrations in these samples were determined by BrCl oxidation, $SnCl_2$ reduction, purge, gold trap, and cold vapor atomic fluorescence spectrometry (CVAFS) detection according to USEPA Method 1631 (USEPA, 1999). For Me-Hg analysis, prepared hair and rice samples were digested using the KOH–methanol/solvent extraction technique (Liang et al., 1994, 1996), and were measured by aqueous ethylation, purge, trap, and gas chromatography-cold vapor atomic fluorescence spectrometer (GC-CVAFS) detection according to USEPA Method 1630 (USEPA, 2001).

Urinary parameters (including urinary pH, protein, and hematuria) were assayed by test paper upon sample collection. Urinary creatinine (U-Cr) contents were analyzed with a HITACHI 7170A automatic analyzer. In order to take hydration and urinary flow rate into account, the assessment of U-Hg as a biomarker for exposure to Hg vapor was adjusted by creatinine excretion. The results of U-Hg were given in μg/g creatinine (μg/g Cr).

2.3. Quality control

Quality control consisted of method blanks, blank spikes, matrix spikes, certified reference material, and blind duplicates. The average T-Hg concentration of hair certified reference material (NIES-13) was 4.30 ± 0.17 μg/g ($n=6$), which was comparable with the certified value of 4.42 ± 0.40 μg/g; the average Me-Hg concentration of 3.84 ± 0.11 μg/g ($n=5$) was in good agreement with the certified value of 3.80 ± 0.40 μg/g. The average T-Hg concentration of rice certified reference material (GBW10010) was 5.4 ± 0.4 ng/g ($n=8$), which is comparable with the certified concentration of 5.3 ± 0.5 ng/g. The recoveries from spiked samples ranged from 80.2% to 111% for Me-Hg in rice sample, and ranged from 98% to 103% for U-Hg. The relative standard deviation was lower than 5% for T-Hg in hair, urine and rice duplicate samples and was lower than 10% for Me-Hg in hair and rice duplicate samples (ranging from 0.8% to 9.9%).

2.4. Statistical method

Statistical analyses were performed by the package of SPSS 11.5 for windows. The characteristics of T-Hg and Me-Hg concentrations were described in Mean ± SD (standard deviation) and examined using descriptive statistics. Mean values were compared between different groups using independent-samples t-test to evaluate the differences. The correlation coefficients among U-Hg, hair T-Hg, and Me-Hg in each group were examined by the Pearson correlation analysis. The results of a statistical test were considered statistically significant if $p < 0.05$.

3. Results

3.1. Hg in urine

Urine, hair Hg concentration, and other parameters were compared between the workers who were involved in artisanal Hg mining and the residents from GX and LWC, and the results are summarized in Table 1. There was no significant difference for both U-Cr and U-pH for different groups. However, significant differences were observed in mean U-Hg between the workers and residents in GX and LWC. The geometric means of U-Hg were 216 and 560 μg/g Cr for the workers in GX and LWC, respectively. The geometric means of U-Hg were 24.8 and 33.1 μg/g Cr for the residents in GX and LWC, respectively. The maximum U-Hg concentration for occupational workers recommended by WHO (1991) was 50 μg/g Cr and that for general population was 5 μg/g Cr (UNIDO, 2003). The highest U-Hg concentration for the workers reached 1940 μg/g Cr, which is about 39 times higher than the occupational exposure limit level (50 μg/g Cr). All the workers' U-Hg concentrations exceeded the limit value, indicated that the workers, which were involved in the artisanal Hg mining in Tongren area, were seriously exposed to Hg vapor. 92.3% of the residents in GX and 100% of the residents in LWC exceeded the limit U-Hg value for general population (5 μg/g Cr), while 30.8% of the residents in GX and 58.3% of the residents in LWC exceeded the limit U-Hg value for the occupational population (50 μg/g Cr). The results indicated that the residents in GX and LWC also were exposed to Hg vapor in a certain level.

3.2. Hg in hair

Concentrations of T-Hg and Me-Hg in hair samples are summarized in Table 1. High hair T-Hg concentrations were found in the workers and the means were 25.5 and 92.0 μg/g for the workers in GX and LWC, respectively. The means of hair T-Hg concentration

Table 1
Urine, hair Hg concentration, and other parameters for the study population.

	GX		LWC	
	Workers ($n=12$) Mean ± SD (range)	Residents ($n=13$) Mean ± SD (range)	Workers ($n=6$) Mean ± SD (range)	Residents ($n=12$) Mean ± SD (range)
Age/year	50 ± 14 (31–76)	64 ± 8 (54–80)	42 ± 5 (33–49)	59 ± 14 (33–78)
U-pH	5.6 ± 0.7 (5.0–7.0)	5.3 ± 0.5 (5.0–6.0)	5.3 ± 0.5 (5.0–6.0)	5.7 ± 0.5 (5.0–6.0)
U-Cr/g L^{-1}	1.17 ± 0.48 (0.47–2.02)	1.34 ± 0.48 (0.45–1.99)	1.33 ± 0.41 (0.72–1.94)	1.34 ± 0.49 (0.78–2.19)
U-Hg/μg L^{-1}	399 ± 415 (50.7–1380)*** 234 (GM)	49.5 ± 37.6 (4.14–95.1) 30.6 (GM)	1130 ± 1110 (150–2820)*** 714(GM)	89.5 ± 92.9 (14.1–365) 39.2(GM)
U-Hg/μg g^{-1}Cr	347 ± 398 (69.4–1430)** 216 (GM)	40.0 ± 37.9 (3.51–137) 24.8 (GM)	917 ± 817 (100–1940)*** 560 (GM)	66.2 ± 50.0 (10.4–167) 33.1 (GM)
Hair T-Hg/μg g^{-1}	25.5 ± 30.7 (4.8–108)** 15.7 (GM)	7.55 ± 2.63 (3.72–12.1)	92.0 ± 116 (21.5–326)** 57.2(GM)	8.26 ± 3.16 (3.52–13.1)
Hair Me-Hg/μg g^{-1}	5.05 ± 2.81 (2.35–10.6)*	3.53 ± 1.19 (1.87–5.65)	5.63 ± 2.99 (2.54–9.55)	4.01 ± 2.20 (2.29–9.23)
Hair Me-Hg ratio/%	37.6 ± 23.5 (4.73–74.1)	50.4 ± 20.2 (23.6–99.3)	10.2 ± 5.09 (2.93–14.7)*	49.9 ± 17.5 (24.0–82.1)

GM: geometric mean.

*** $p<0.001$ when compared with the residents in the same site.
** $p<0.01$ when compared with the residents in the same site.

Table 2
The correlation coefficients among the concentration of different Hg speciation for the workers and residents group.

		U-Hg/μg g^{-1}Cr	Hair T-Hg	Hair Me-Hg	Hair Me-Hg ratio
Workers	U-Hg/μg L^{-1}	0.93***	0.74***	0.48*	−0.26
	U-Hg/μg g^{-1}Cr		0.65**	0.53*	−0.27
	Hair T-Hg			0.40	−0.51*
	Hair Me-Hg				0.14
Residents	U-Hg/μg L^{-1}	0.80***	0.18	−0.17	−0.41*
	U-Hg/μg g^{-1}Cr		0.38	−0.02	−0.46*
	Hair T-Hg			0.57**	−0.39
	Hair Me-Hg				0.49*

*** $p<0.001$
** $p<0.01$.
* $p<0.05$.

were 7.55 and 8.26 μg/g for the residents in GX and LWC, respectively. Significant differences ($p<0.01$) were observed in the means of hair T-Hg concentrations between the workers and residents in GX and LWC, respectively. Hair T-Hg analysis is difficult in differentiating between exogenous metal contamination and the metal deposited endogenously (Hat and Krechniak, 1993), since elemental Hg may adhere to hair and is hard to be removed by washing steps, especially for the occupational population associated with serious external Hg exposure (Li et al., 2008d). A significant correlation was observed between hair I-Hg and TGM in ambient air in Wuchuan Hg mining area (Li et al., 2008c), which indicated that hair T-Hg concentration can be impacted by the exposure to Hg vapor.

However, hair still can be used as an indicator for Me-Hg exposure, because generally there is almost no exogenous contamination of Me-Hg in hair (Li et al., 2008d). Therefore, both hair T-Hg and Me-Hg concentrations were measured to distinguish the Me-Hg intake and external exposure to Hg vapor. The means of hair Me-Hg concentrations for the workers and residents in GX were 5.05 and 3.53 μg/g, respectively and the corresponding values were 5.63 and 4.01 μg/g for the workers and residents in LWC, respectively. The highest hair Me-Hg concentration in the study population reached 10.6 μg/g. This value is close to the threshold for the onset of symptoms for Me-Hg, which is recognized to be 10–14 μg/g in maternal hair (NRC, 2000). The hair Me-Hg concentrations were significantly elevated compared with the value of 0.65 μg/g from the control group in Changshun, Guizhou Province (Li et al., 2008b), which indicated the Me-Hg exposure for the population in the study area.

3.3. The relation between urine and hair Hg

The correlation coefficients between U-Hg, hair T-Hg, hair Me-Hg, and hair Me-Hg ratio for the workers and residents groups are listed in Table 2. For the residents, hair Me-Hg shows significant correlation ($r=0.57$, $p<0.01$) with hair T-Hg, which is consistent with most studies. On the contrary, hair T-Hg concentrations had no significant correlation with hair Me-Hg concentrations, but had a significant correlation with U-Hg ($p<0.01$) for the workers. Moreover, Me-Hg concentrations in hair samples just accounted for 37.6% and 10.2% of T-Hg on average for the workers in GX and LWC, respectively, which is much lower than the residents. T-Hg and the Me-Hg ratio in the hair samples showed significant negative correlations ($r=-0.49$; $p<0.01$; Fig. 1). This indicated different exposure pathways for Hg vapor and Me-Hg and the workers were exposed to Hg vapor through inhalation (Li et al., 2008b, c).

3.4. Hg in rice and its relation with hair Hg

The concentrations of T-Hg and Me-Hg in the rice samples collected from the study area are listed in Table 3. The average of T-Hg concentrations were 134 and 138 ng/g in the rice collected from GX and LWC, respectively. The concentrations of T-Hg in all rice samples exceeded the national guidance limit for foodstuff other than fish, which is 20 ng/g recommended by the Chinese National Standard Agency (CNSA, 1994). The average of Me-Hg concentrations were 15.4 and 14.4 ng/g in the rice collected from GX and LWC, respectively, and Me-Hg constituted of 14.8 ± 9.57% (4.17–40.0%)

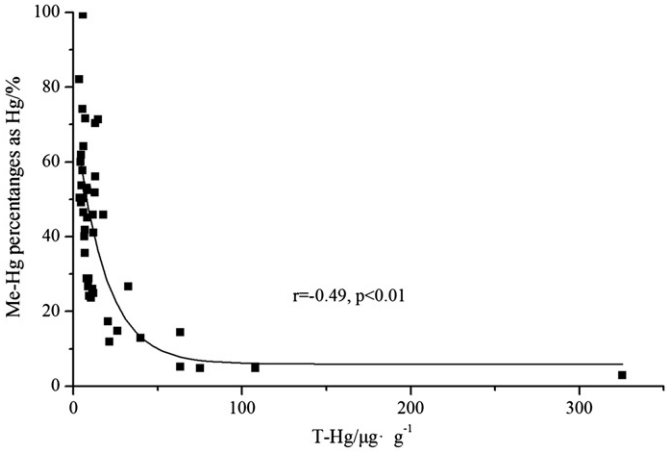

Fig. 1. The relationship between T-Hg levels and Me-Hg percentages as Hg in the hair for the study population.

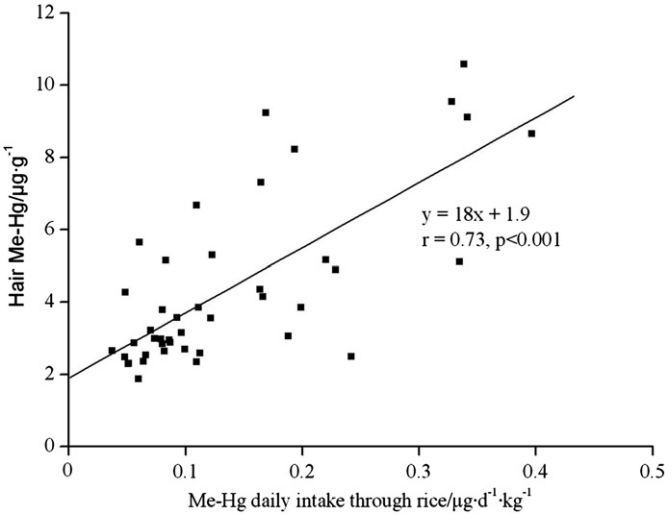

Fig. 2. Correlation between hair Me-Hg concentrations and Me-Hg daily intake from rice for the study population.

Table 3
T-Hg and Me-Hg concentrations in the rice from the study area.

		Min	Max	Mean	SD	*n*	Distribution pattern
GX	T-Hg/ng g^{-1}	40.9	277	134	79.7	25	Normal
	Me-Hg/ng g^{-1}	6.81	33.5	15.4	7.39	25	Normal
	Me-Hg ratio/%	4.17	40.0	14.8	9.57	25	Normal
LWC	T-Hg/ng g^{-1}	29.5	258	138	64.1	18	Normal
	Me-Hg/ng g^{-1}	6.37	34.1	14.4	9.75	18	Normal
	Me-Hg ratio/%	5.21	63.5	9.88[a]	13.5	18	Log normal

[a] Geometric mean.

Table 4
Daily Me-Hg intake through rice consumption for the study population/μg d^{-1} kg^{-1}.

Site	Min	Max	Mean	SD	*n*
GX	0.048	0.396	0.148	0.098	25
LWC	0.037	0.341	0.122	0.088	18

and 9.88 ± 13.5% (5.21–63.5%) for T-Hg in rice samples collected from GX and LWC, respectively. No significant correlation ($r=0.28$, $p=0.07$) was found between the T-Hg and Me-Hg concentrations in the rice from the study area. Our results were comparable to other studies obtained in rice samples in Hg mining areas in Guizhou, China (Horvat et al., 2003; Feng et al., 2008; Li et al., 2008c; Qiu et al., 2008).

In order to identify the Me-Hg exposure source, we estimated the daily intake of Me-Hg for of the study population. In this study, we selected those residents who stayed in the study areas during the previous 3 months and always consumed rice planted by themselves to evaluate their Me-Hg exposure. According to the daily rice consumption data provided by the participants, and Me-Hg concentrations in rice, daily Me-Hg intakes for the population in the study area were calculated (Table 4). A significant correlation ($r=0.73$, $p<0.001$) between hair Me-Hg concentrations and daily Me-Hg intakes from rice for the study population was observed as shown in Fig. 2. This confirmed that rice intake was the main Me-Hg exposure route for study population in Tongren Hg mining area, which was consistent with our previous study (Feng et al., 2008).

Table 5
Clinical symptoms and correlative parameters for the Hg poisoning workers.

	Site	Clinical symptom	U-Hg/ μg g^{-1} Cr	Hair Me-Hg/ μg g^{-1}
Worker 1	GX	Lightly finger tremor	107	8.65
Worker 2	GX	Lightly finger tremor	234	2.96
Worker 3	LWC	Lightly finger tremor	100	2.54

3.5. Clinical symptoms

For Hg vapor exposure, when U-Hg level is above 100 μg/g Cr, the probability of developing the classical neurological signs of Hg intoxication (tremor, erethism) and proteinuria is high (WHO, 1991). The geometric means of U-Hg were 216 and 560 μg/g Cr for the workers in GX and LWC, respectively. Most of the workers' U-Hg levels exceeded 100 μg/g Cr; therefore, the workers were at a high risk of Hg vapor poisoning. For the Me-Hg exposure, hair Hg threshold for showing onset of neurological symptoms in human body is 50 μg/g (WHO, 1990). Therefore, the study population is not at risk for clinical neurological disorders.

Clinical symptoms and correlative parameters for the Hg poisoning workers in GX and LWC are listed in Table 5. Symptoms (finger tremor) were observed in three workers. These indicated that physical impairments occurred due to serious exposure to Hg vapor during artisanal Hg mining.

4. Discussions

The comparison of human U-Hg concentration between this study and other data from Hg mining areas over the world is shown in Table 6. The U-Hg levels of the artisanal mining workers in the study area were highly elevated when compared with other studies got from other Hg mines, and were equal to the results from artisanal mining workers in Wuchuan. The documented maximum value was 10565 μg L^{-1}, which obtained from the workers in Abbadia San Salvatore mine, in the period 1968–1983. Physical impairments and clinical symptoms were observed in three workers, although their U-Hg concentrations were not above those of the study population. The U-Hg reflects short term exposure and the observed effects may result from long-term exposure.

Table 6
The comparison of human U-Hg concentrations in Hg mining areas over the world.

Location	*n*	Mean ± SD ($\mu g\ g^{-1}$ Cr)	Range ($\mu g\ g^{-1}$ Cr)	Comments	Refs.
Idrija, Slovenia	54	69.3 ± 31.4	26–158	Mercury miners, average annual past exposure U-Hg levels	Kobal et al. (2004)
Algeria	64	139 ± 80.9	33–382	Workers from the mercury production plant	Abdennour et al. (2002)
Monte Amiata, Italy	606	160 $\mu g\ L^{-1}$ AM 83 $\mu g\ L^{-1}$ GM	1.3–10565μgL^{-1}	Workers in Abbadia San Salvatore mine, in the period 1968–83	Bellander et al. (1998)
Almadén, Spain		48–797 $\mu g\ L^{-1}$	11–2194μgL^{-1}	Different jobs in the period of 1972–1986	Garcia-Gomez et al. (2007)
Wuchuan, China	22	1060 ± 1510 463 (GM)	28–6150	Mercury smelting workers	Li et al., 2008b
Wanshan, China	35	56.9	2.4–291.3	Local residents, and some may be involved in artisanal Hg mining	Feng et al. (2008)
GX, Tongren, China	12	347 (AM) 216 (GM)	69.4–1430	Artisanal Hg mining workers	This study
	13	40.0 (AM) 24.8 (GM)	3.51–137	Local residents	
LWC, Tongren, China	6	917 (AM) 560 (GM)	100–1940	Artisanal Hg mining workers	This study
	12	66.2 (AM) 33.1(GM)	10.4–167	Local residents	

Table 7
The comparison of hair Hg concentration from different studies worldwide.

Location	Population	Hair T-Hg level	Exposure source	Refs.
New Zealand	Mother–child pairs	8.3 (6.0–21)	Fish	Kjellstrom et al. (1986, 1989), Crump et al. (1998)
Faroe Islands	Pregnancy women	4.3 (2.6–7.7) 12.5	Pilot whale	Grandjean et al. (1997, 1998)
Seychelles	Mother–infant pairs	5.9 (0.5–26.7)	Fish	Marsh et al., 1995; Myers et al., 1995, 2003
Five coastal cities in China	General population	0.83 (0.04–8.48)	Fish	Liu et al. (2008)
Zhoushan Island, China	General population	Fathers 3.8 (0.9–9.5)[a] Mothers 1.8 (0.3–4.1)[a] Children 1.7 (0.6–4.1)[a]	Fish	Cheng et al. (2009)
Amazon	Indigenous and riparian population Urban residents	≈ 15 ≈ 2–3	Fish	Numerous studies, reviewed by Passos and Mergler (2008)
Nine prefectures in Japan	General population	Male 2.42 (0.10–29.4) Female 1.37 (0.02–25.8)	Fish	Yasutake et al. (2004)
Songhua River, Northeast China	Fishmen	2.25 (0.02–18.1)	Fish	Qiu et al. (1994)
Wanshan Hg mining area, China	Local residents	2.8 (0.8–5.6)[a] 1.3 (0.2–3.6) [a] 1.5 (0.5–3.5)[a]	Rice	Feng et al. (2008)
Wuchuan Hg mining area, China	Local residents	1.38 (0.45–5.89)[a]	Rice	Li et al., 2008c
Tongren Hg mining area, China	Workers and local residents	4.26 (1.87–10.6)[a] 4.55 (2.29–9.55)[a]	Rice	This study

[a] Me-Hg.

The means of U-Hg were 40.0 and 66.2 $\mu g\ g^{-1}$ Cr for the residents in GX, LWC, which was in the same level in the residents in Wanshan Hg mining areas (Feng et al., 2008). But the U-Hg levels were close to 50 $\mu g\ g^{-1}$ Cr for occupational exposure limit, also indicated a certain level of Hg vapor exposure.

The comparison of hair Me-Hg concentration between this study and other studies worldwide is shown in Table 7. From the comparison, we found that the population in the study area had the high hair Me-Hg concentrations among the studies in China, even compared with fish eating population in the costal and island area. China is considered as the largest source of anthropogenic Hg emissions, but the Me-Hg concentrations in fish tissue in China were very low, even in the heavily Hg contaminated area in Guizhou Province, for instance, the Baihua Reservoir (Yan et al., 2008), and Wanshan Hg mining area (Qiu et al., 2009). The low level of Hg in fish may be due to the fast growth of the cultivated fish and the shortness and simple of the aquatic food chain (Yan et al., 2008).

Low fish Hg concentration resulted in low hair Hg levels in the fish eating population in China. Low average hair T-Hg concentrations (< 1 $\mu g/g$) were reported in fish eating population in five coastal sites (Shanghai, Ningbo, Dalian, Xiamen, and Zhoushan) in eastern China (Liu et al., 2008), and Di'er Songhua River, northeastern China (Zhang and Wang, 2006).

Even so, the hair Hg levels from our studies were much lower than that from the studies in Amazon, New Zealand, Faroe Islands, and Seychelles. But some value was close to the onset of neurological symptoms in maternal hair recommended both by WHO and the NRC, 2000 (10–14 $\mu g/g$), the health risk to neonates from prenatal Me-Hg exposure may still exist.

Generally humans are exposed to Me-Hg through contaminated fish and marine mammals, but recent studies confirmed the risk of Me-Hg from rice consumption. The risk assessment indicated the daily Me-Hg intake through rice consumption for some population in GX and LWC exceeded the JECFA recommended value of

0.23 μg d^{-1} kg^{-1} (JECFA, 2003) and the daily Me-Hg intake for the majority of the residents in Hg mining area Guizhou Province, China exceeded the USEPA recommended value of 0.10 μg d^{-1} kg^{-1} (USEPA, 1997). Therefore, the residents in GX and LWC were at a high health risk of Me-Hg exposure and the residents in other Hg mining areas were also at a potential health risk of Me-Hg exposure through rice intake.

This study shows human co-exposure to Hg vapor and Me-Hg through different routes. There are no data on the combined exposure to Hg vapor and Me-Hg exposure for human, despite concerns that combined exposure may elevate risk via additive or synergistic mechanisms. Additional research is clearly needed to address this question.

5. Conclusions

The results confirmed co-exposure to Hg vapor and Me-Hg for the population in GX and LWC, Tongren artisanal Hg mining area, Guizhou Province. The workers, who were involved in artisanal Hg mining, showed extremely high U-Hg levels and some neurobehavioral impairments occurred due to the serious Hg vapor exposure. The local residents were also exposed to Hg vapor in a certain level. And the whole population was at risk of Me-Hg exposure through rice consumption.

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