Environmental Research 140 (2015) 198–204

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

# Human inorganic mercury exposure, renal effects and possible pathways in Wanshan mercury mining area, China

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ARTICLE INFO

*Article history:*
Received 16 January 2015
Received in revised form
26 March 2015
Accepted 29 March 2015
Available online 9 April 2015

*Keywords:*
Inorganic mercury
Human exposure
Renal effect
Pathway

ABSTRACT

Rice can accumulate methylmercury (MeHg) and rice consumption is the main route of MeHg exposure for the local population in Guizhou, China. However, inorganic Hg (IHg) load in human body is not comprehensively studied in highly Hg polluted areas such as Hg mining areas. This study is designed to evaluate human IHg exposure, related renal effects and possible pathways in Wanshan Hg mining area, Guizhou, Southwest China. Residents lived within 3 km to the mine waste heaps showed high Urine Hg (UHg) concentrations and the geometrical means (Geomean) of UHg were 8.29, 5.13, and 10.3 μg/g Creatinine (Cr) at site A, D, and E, respectively. It demonstrated a gradient of UHg concentrations with the distance from the pollution sources. A significantly positive correlation between paired results for UHg concentrations and serum creatinine (SCr) was observed in this study, but not for UHg and blood urea nitrogen (BUN). There are significant increases of SCr in two quartiles with high UHg concentrations. The results indicated that human IHg exposure may cause impairment of renal function. By calculation of Probable Daily Intake from different routes, we found that dietary intake is the main pathway of IHg exposure for the local population, rather than inhalation of Hg vapor.



## 1. Introduction

Mercury (Hg) and its compounds are recognized as potentially hazardous materials and are rated in the top category of environmental pollutants. Mercury can cause significantly adverse effects on human health. The toxicity of mercury depends on its chemical forms. Inhalation of mercury vapor via dental amalgam filling and ingestion of methylmercury (MeHg) via fish consumption are the most important routes of human exposure to mercury (WHO, 1990, 1991). For the occupational exposure (such as workers in chlor-alkali plants, Hg mines, Hg based gold extraction, Hg processing and sales, thermometer factories, and dental clinics), inhalation of Hg vapor is the most important route of human exposure to Hg.

About 80% of the inhaled Hg vapor is retained in the bloodstream and from there is distributed to the tissues. Urine and feces are the principal routes of mercury elimination and the urinary route dominates when exposure is high (WHO, 1991). The half-life for Hg in urine is about 2 months. Urine Hg (UHg) measurements are widely used for assessment of inorganic Hg (IHg) exposure in humans because UHg is thought to be indicating most closely the mercury levels present in the kidneys (Clarkson et al., 1988; Barregard, 1993).

Neurological effects and renal effects have been observed following exposure to elemental Hg (WHO, 1991; UNEP, 2002). Limited epidemiological studies are available concerning neurological and renal toxicity following oral exposure to IHg (USEPA, 1997; ATSDR, 1999). Since inhaled elemental Hg can be oxidized into IHg, it is indistinguishable from IHg from digestion sources (USEPA, 1997). The major effect from chronic exposure to elemental Hg and IHg is kidney damage. To evaluate the impact of occupational Hg exposure on renal function, glomerular function is commonly estimated by high-molecular-weight proteins in urine (such as albumin (ALB), transferin and IgG), whereas tubular function is assessed by low-molecular-weight proteins in urine (such as α1-microglobulin (α1-MG), β2-microglobulin (β2-MG) and retinol binding protein (RBP)) and the activity of some tubular enzymes (N-acetyl-β-D-glucosaminidase (NAG)) (Franko et al., 2005; Langworth et al., 1992; Cárdenas et al., 1993; Al-Saleh et al., 2012). As well, serum creatinine (SCr) and blood urea nitrogen (BUN) are most commonly indicators of renal function, which also can be used as markers of nephrotoxic assessment on environmental exposures to Hg (Li et al., 2013).

Mercury mining areas are considered as the hot spots of Hg

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http://dx.doi.org/10.1016/j.envres.2015.03.033
0013-9351/

pollution and the impact from Hg mining activities in Guizhou Province, China are widely investigated (Feng and Qiu, 2008). Occupational exposures to Hg vapor (Li et al., 2008, 2011a) and MeHg bio-accumulation in the food chain and related human health risk assessments (Feng et al., 2008; Zhang et al., 2010a) in Guizhou Hg mining areas gained much concerns. As well, the Hg concentrations in ambient air were highly elevated with a range of two orders of magnitude (17–2100 ng/m$^3$) in Wanshan area even after the closure of Hg mines (Dai et al., 2012). THg concentrations in crops cultivated in Hg mining areas were highly elevated when compared with the National Permitted Limit of 20 μg/kg in foods. It indicated that the general populations in Wanshan Hg mining area may exposure to IHg through inhalation of Hg vapor and dietary intake. For fish eating population, the positive relation was also found between fish consumption and IHg in both blood and urine, which may result from absorption of IHg from fish or from demethylation of MeHg (Passos et al., 2007).

This study is designed to evaluate human IHg exposure and related renal effects in Wanshan Hg mining area. Efforts were also made to identify the possible pathway of IHg exposure for the general population, which can provide scientific data for risk assessment and risk control of human Hg exposure in Hg polluted areas. In a companion paper, human MeHg exposure and the relationship between human body burden and dietary MeHg intake are discussed (Li et al., 2015).

## 2. Materials and methods

### 2.1. Study area

Wanshan Hg mining area is selected for this study. It is located in the eastern part of Guizhou Province and it was the largest Hg mine in China. Large scale mining activities officially ceased in 2001. The long history of mining activities has resulted in serious Hg contamination to the local environment. Wanshan County is consisted of 5 towns, namely Wanshan, Huangdao, Xiaxi, Aozhai, and Gaolouping. Seven sites (A–G) in Xiaxi and Aozhai Town were selected in this study, which locate along the Aozhai and Xiaxia River with different distances from the Hg pollution sources (Fig. 1).

### 2.2. Samples collection

Sampling was conducted in December 2012. The local residents staying at home over 3 months were recruited in this study. The recruitment strategy in the survey was to recruit 10–20% of whole population at each site. All the participants are voluntarily participated in this study. The recruitment period lasts two days. A questionnaire was also conducted to obtain the basic information on age, body weight, profession, history of involvement of artisanal Hg mining activity, dental fillings, smoking and alcohol drinking habits, illness, and the amount of daily rice consumption. In order to evaluate the effect of IHg exposure on renal function, the participants with known nephropathy or other serious illnesses are not included in this study.

Urine samples were collected in clean plastic centrifugal tubes, hermetically sealed and stored at 4 °C until analysis. Approximately 5 ml venous blood samples were collected from each participant using a metal-free plastic vacutainer without anticoagulant, which was centrifuged immediately at 3000 rpm for 10 min to obtain the serum.

The present study obtained ethics approval from the Institute of Geochemistry, Chinese Academy of Sciences. All participants were required to sign a consent form.

### 2.3. Analytical methods

Urine samples were digested in a water bath (95 °C) with a fresh mixture of $HNO_3/H_2SO_4$ (v/v 4:1) for THg analysis. The digests were determined by BrCl oxidation, $SnCl_2$ reduction, purge, gold trap, and cold vapor atomic fluorescence spectrometry (Yan

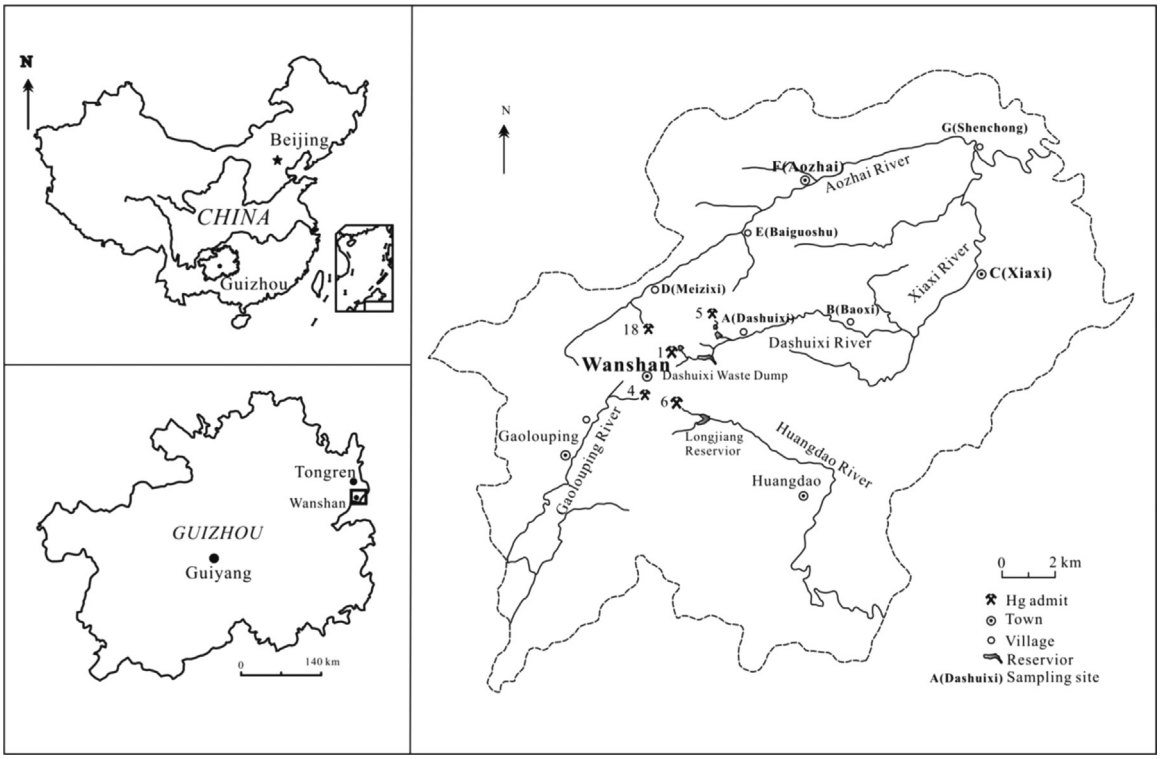


**Fig. 1.** Location of sampling sites in the study area.

et al., 2005).

Urine creatinine (UCr), serum creatinine (SCr) and blood usea nitrogen (BUN) were analyzed with automatic biochemical analyzer in Wanshan Hospital. In order to take hydration and urinary flow rates into account, the assessment of UHg as a biomarker for exposure to IHg was adjusted by creatinine excretion. Its results were given in the unit of μg/g creatinine (μg/g Cr).

Blood and hair IHg concentrations were obtained by the difference between THg and MeHg concentrations. THg concentrations in the hair samples were analyzed by the RA-915+ Hg analyzer coupled with PYRO-915+ attachment (Lumex, Russia). THg in blood samples were acid digested and determined by cold vapor atomic fluorescence spectrometry. Hair and blood MeHg were digested using KOH-methanol/solvent extraction technique (Liang et al., 1996) and then measured using aqueous ethylation, purge, trap, and GC-CVAFS detection (Brooks Rand MERX).

Total gaseous mercury (TGM) concentrations in indoor and outdoor air of different sites were measured in situ by a portable atomic absorption spectrometer with Zeeman background correction (Lumex RA-915+, Russia). The detection limit of the instrument is as low as 2 ng/m$^3$. It can provide the real-time analysis with a response time of 1 s. A single concentration data represents an average value of a time interval of 10 s and the measurements at each site were carried out at least for 10 min.

Quality control system consists of method blanks, blank spikes, matrix spikes, certified reference material (CRM) and blind duplicates. Limit of determination (LOD) was 0.01 μg/L for UHg. The results of certified reference materials are listed Table 1. The relative percentage difference was lower than 10% for UHg in duplicate samples.

### 2.4. Calculation of probable daily intake (PDI)

We carried out the investigation of IHg exposure via drinking water, inhalation, and food consumption at site A. The PDI values for general adult population were calculated according to the following formula:

$$\mathrm{PDI} = \Sigma(C_i \times \mathrm{IR}_i \times A_i)/\mathrm{bw} \quad (1)$$

where PDI is given in micrograms per kilogram of body weight per day (μg/kg/d); bw (body weight)=60 kg; $C$ is IHg concentration; IR is daily intake rate; $A$ is absorption rate.

### 2.5. Data analysis

All data were analyzed using IBM SPSS 19 for windows. The data are tested for normal distribution by the Kolmogorov–Smirnov test. If they are not normally distributed, the data are log transformed for further statistical analysis. The characteristics of the data were described in Mean ± Standard Deviation (SD) and Geomean for descriptive statistics. Mean values of the data at different sites were compared using ANOVA. Relationships between UHg, hair and blood IHg, UHg and BUN, SCr are analyzed using the Pearson correlation analysis. Results of statistical tests were considered statistically significant if $p < 0.05$.

**Table 1**
List of certified reference materials used in the present study and the results obtained. Data obtained are reported in italics and in brackets as a mean value and standard deviation.

| Producer | CRM | Matrix | Certified value (Mean ± SD) | | |
|---|---|---|---|---|---|
| | | | THg | MeHg | Unit |
| CCDC | ZK020-2 | Urine | 45 ± 4.0 *(46 ± 4.2)* | | μg/L |
| Japan NIES | NIES-13 | Human hair | 4.4 ± 0.2 *(4.3 ± 0.09)* | 3.8 ± 0.4 *(3.74 ± 0.24)* | μg/g |
| CCDC | ZK021-2 | Bovine blood | 30 ± 3 *(31 ± 2.4)* | | μg/L |

NIES: National Institute for Environmental Studies; CCDC: Chinese Center for Disease Control and Prevention.

## 3. Results and discussion

### 3.1. IHg exposure

Basic information of the study population at different sites is listed in Table 2. As expected, most of recruited participant numbers in the survey accounted 10–20% of whole population at each site except site C and F, which are big town with large population. Data of UHg, hair and blood IHg are summarized in Table 3. There was no significant difference of UCr in different groups. However, the mean UHg at site A, D, and E were significantly higher than these at other sites, while site C and G can be considered as the control site in this study. The maximum UHg concentration for the occupational workers recommended by the WHO (1991) was 50 μg/g Cr, while UHg levels rarely exceed 5 μg/g Cr in people who are not occupationally exposed to Hg (UNIDO, 2003). The geomean of UHg at site A, D, and E were 8.29, 5.13 and 10.3 μg/g Cr, respectively, which were considerably higher than the value of 1.17 μg/g Cr at site G and the value of 5 μg/g Cr set for the general population. The highest UHg in this study reached 144 μg/g Cr, which is about 2 times higher than the occupational exposure limit (50 μg/g Cr) recommended by WHO (1991) and 2.5% of the investigated population (4/160) had UHg exceeding this limit. Thirty-six percent of the investigated population (57/160) had UHg exceeding the general value recommended by UNIDO (5 μg/g Cr), which indicated that the study population was exposed to IHg in high levels.

The UHg concentrations at site A, D, and E were comparable to that in XCX from our previous study (7.2 μg/g Cr; Feng et al., 2008) and significantly lower than these of occupational workers involved in artisanal Hg mining in Wuchuan (463 μg/g Cr; Li et al., 2008) and Tongren (216 and 560 μg/g Cr for GX and LWC; Li et al., 2011a). The UHg at site B, F, and G (with the average of 1.07, 1.29 and 1.48 μg/g Cr) basically represented the regional background level, which were comparable to that of the control group in Changshun (1.3 μg/g Cr; Li et al., 2008).

With regard to the site difference of UHg concentrations, it demonstrated a gradient at different sites with the distance to the pollution sources in the two catchments (Fig. 2). The significant elevated UHg levels were observed in the upstream region (site A, D, and E), which were seriously impacted by historical Hg mining activities (Fig. 2). The UHg levels at site C and G basically represented the regional background levels, which were not significantly impacted from Hg mining activities; and THg concentrations in natural soil of site C and G were less than 0.5 mg/kg (Dai et al., 2013).

Similar with the spatial distribution of UHg concentrations, the averages of blood and hair IHg at different sites also demonstrated a gradient with the distance to the pollution sources in the two catchments (Table 3). As well, UHg had significant correlations with hair IHg and blood IHg ($p < 0.001$). It confirmed that hair and blood IHg might be a useful tool for monitoring IHg exposure and is consistent with our previous study (Li et al., 2011b).

### 3.2. Renal effects

Statistical results of parameters of renal effects are listed in

**Table 2**
Basic information of the study population.

| Site | Name | Population | *n* | Male | Female | Age (years) | Height (cm) | Weight (kg) |
|---|---|---|---|---|---|---|---|---|
| A | Dashuixi | 100 | 32 | 14 | 18 | 51.7 ± 13.0 | 158 ± 8.3 | 54.5 ± 9.0 |
| B | Baoxi | 150 | 13 | 3 | 10 | 43.9 ± 13.8 | 156 ± 7.4 | 56.2 ± 9.9 |
| C | Xiaxi | 2000 | 28 | 11 | 17 | 39.5 ± 11.7 | 160 ± 6.6 | 59.7 ± 9.3 |
| D | Meizixi | 150 | 28 | 13 | 15 | 52.4 ± 16.7 | 155 ± 8.4 | 51.3 ± 10.9 |
| E | Baiguoshu | 150 | 26 | 12 | 14 | 50.4 ± 11.0 | 157 ± 8.4 | 55.4 ± 8.6 |
| F | Aozhai | 1500 | 11 | 6 | 5 | 50.6 ± 13.8 | 159 ± 6.6 | 55.2 ± 9.1 |
| G | Shenchong | 200 | 22 | 13 | 9 | 52.3 ± 14.8 | 159 ± 6.9 | 58.7 ± 12.7 |
| Total | | 4250 | 160 | 72 | 88 | 48.9 ± 14.3 | 158 ± 7.8 | 55.9 ± 10.3 |

Table 4. The typical human reference ranges for SCr are 45–90 μmol/L for women and 60–110 μmol/L for men, respectively. Normal range of BUN for adults is 1.7–8.3 mmol/L by this determination method. There are only 1 woman exceeded the SCr limit and 17 adults exceeded the BUN limit (10.6%). Even though we got rid of the participants with known nephropathy or other serious illnesses, the study population revealed a high BUN level. There is no difference of UHg between population with high BUN levels and with a normal level, which indicated high BUN levels are not related to Hg exposure.

The mean of BUN at site E was significantly higher than these at site B and C and there is no significant difference of SCr levels among different sites (Table 4). However, a significantly positive correlation between paired results for UHg concentrations (μg/g Cr) and SCr (μmol/L) was observed in this study ($p < 0.05$); but no significant correlation between UHg concentrations (μg/g Cr) and BUN (mmol/L) was obtained. To further examine the renal effects of IHg exposure, the study population is divided into four equal groups according to the UHg concentrations. Since sex difference of SCr concentrations, sex distributions in four groups were compared and there is no difference. Then BUN and SCr levels in each quartile were compared (Fig. 3). The SCr levels in the third (Q3) and fourth quartiles (Q4) were significantly higher than those from the first quartile (Q1) ($p < 0.05$ and $p < 0.01$, respectively); but no significant differences was observed for BUN levels among different quartiles. An increase of SCr levels was found in quartiles with the highest UHg concentrations, which indicated that IHg exposure may result in renal impairment.

The results suggest that IHg exposure impairs renal function, since the increase of SCr is a sensitive biomarker of renal disease. SCr is more sensitive than BUN to reveal renal effect of IHg exposure in the study population, which is consistent with a previous study also conducted in abandoned Hg mines (Li et al., 2013). Since SCr is a common test in the local hospital, it can be used as an effective approach for diagnosis of renal effect resulted from IHg exposure.

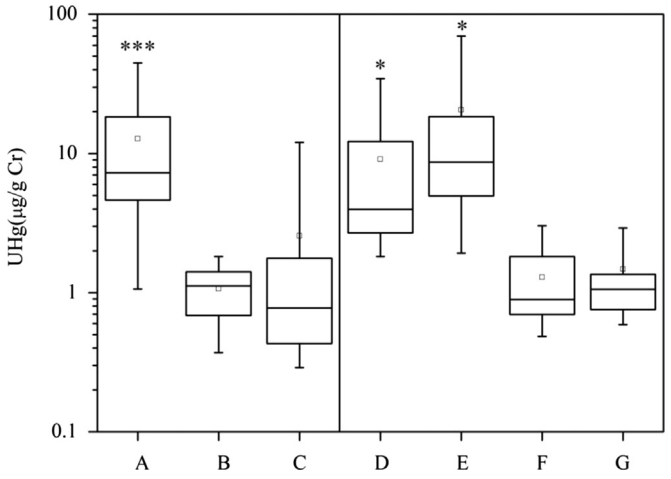


**Fig. 2.** Comparison of UHg concentrations in the population at different sites. ***, $p < 0.001$; *, $p < 0.05$; compared with G. Each box represents interquartile range (25th and 75th percentile), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentile.

### 3.3. Exposure pathways

To evaluate the contribution from inhalation of Hg vapor, total gaseous mercury (TGM) concentrations of indoor and outdoor ambient air were determined in the study area and the results are shown in Fig. 4. TGM concentrations ranged from 13.6 to 344 ng/m$^3$ at different sites, which were 1–2 orders of magnitude higher than those of global background (1–2 ng/m$^3$). Interestingly, TGM concentrations of indoor ambient air were generally higher than those of outdoor ambient air at the same site (except site F), which may be attributed to winter household heating using coal (Feng et al., 2004). With regard to TGM concentrations in ambient air, the results in this study (13.6–344 ng/m$^3$) were comparable with data of 17.8–1101.8 ng/m$^3$ reported by Wang et al. (2007) and those of 17–2100 ng/m$^3$ reported by Dai et al. (2012) which was

**Table 3**
Statistical results of UHg, hair and Blood IHg in the population at different sites (Mean ± SD, (Geomean)).

| Site | *n* | UCr (g/L) | UHg (μg/L) | UHg (μg/g Cr) | Hair IHg (μg/g) | Blood IHg (μg/L) |
|---|---|---|---|---|---|---|
| A | 32 | 1.44 ± 0.57 | 16.2 ± 16.1(10.9)A | 12.8 ± 12.4(8.29)A | 0.81 ± 0.73(0.39) | 6.61 ± 4.31(4.93)a,b |
| B | 13 | 1.08 ± 0.31 | 1.17 ± 0.60(0.99)B | 1.07 ± 0.45(0.97)B | 0.64 ± 0.42(0.56) | 2.75 ± 1.91(1.56)b |
| C | 28 | 1.07 ± 0.38 | 1.62 ± 2.99(0.94)B | 2.56 ± 5.77(0.99)B | 0.56 ± 0.69(0.33) | 2.25 ± 2.47(0.98)b |
| D | 28 | 1.17 ± 0.23 | 11.0 ± 18.0(5.89)A | 9.09 ± 12.9(5.13)A | 1.32 ± 1.61(0.75) | 8.57 ± 5.95(7.07)a |
| E | 26 | 1.26 ± 0.18 | 24.1 ± 29.6(12.8)A | 20.6 ± 30.5(10.3)A | 2.09 ± 5.04(0.46) | 15.6 ± 30.0(7.22)a |
| F | 11 | 1.26 ± 0.23 | 1.63 ± 1.18(1.35)B | 1.29 ± 0.82(1.09)B | 0.72 ± 0.77(0.46) | 5.93 ± 2.47(5.57)a,b |
| G | 22 | 1.22 ± 0.25 | 1.76 ± 1.78(1.38)B | 1.48 ± 1.41(1.17)B | 0.52 ± 0.32(0.36) | 4.93 ± 1.32(4.68)a,b |
| Total | 160 | 1.23 ± 0.37 | 9.82 ± 17.9(3.51) | 8.32 ± 16.1(3.04) | 0.99 ± 2.19(0.45) | 7.05 ± 13.2(3.65) |

There is significant difference between those containing different letters. Upper case letter, $p < 0.01$; lower case letter, $p < 0.05$.

**Table 4**
Statistical results of parameters of renal effects in the population at different sites (Mean ± SD).

| Site | *n* | BUN (mmol/L) | SCr (μmol/L) |
|---|---|---|---|
| A | 32 | 6.00 ± 2.54a,b | 62.9 ± 10.7 |
| B | 13 | 5.22 ± 1.63b | 52.7 ± 11.0 |
| C | 28 | 5.72 ± 1.50b | 57.8 ± 6.54 |
| D | 28 | 6.38 ± 1.80a,b | 63.7 ± 8.25 |
| E | 26 | 6.90 ± 1.92a | 64.4 ± 13.8 |
| F | 11 | 6.59 ± 1.44a,b | 56.2 ± 5.30 |
| G | 22 | 5.96 ± 1.03a,b | 57.9 ± 6.54 |
| Total | 160 | 6.13 ± 1.86 | 60.4 ± 10.1 |

There is significant difference between those containing different letters. Lower case letter, $p < 0.05$.

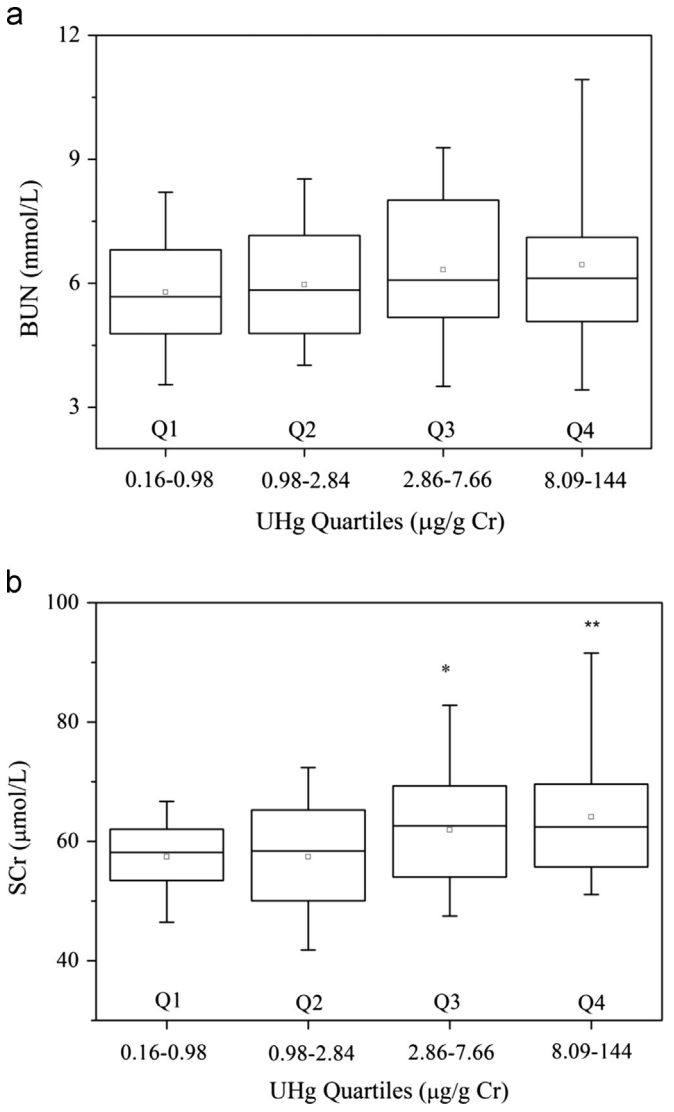


**Fig. 3.** Quartile analysis of BUN and SCr versus UHg in the study population. *, $p < 0.05$; **, $p < 0.01$; compared with Q1.

also carried out in Wanshan area. TGM concentrations in ambient air in this study are below 20 μg/m$^3$ of the Chinese Occupational Criterion (GB16227-1996); but some values exceeded the annual average of Reference Limit of Ambient Air Quality Standards (50 ng/m$^3$; GB3095-2012). The spatial distribution of TGM concentrations in ambient air indicated that the highest levels were

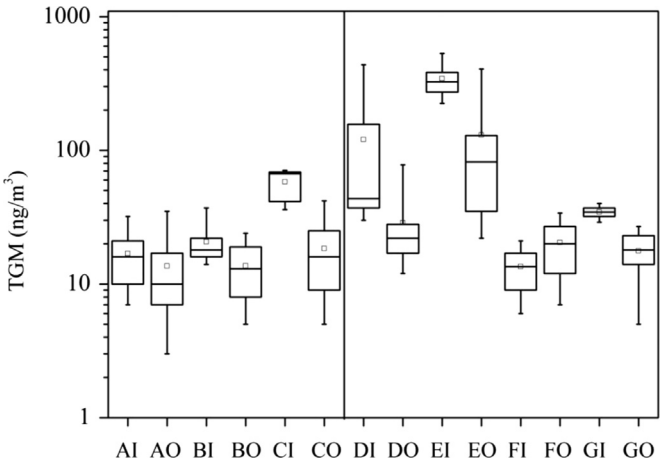


**Fig. 4.** TGM concentrations of indoor and outdoor air at different sites (O, outdoor; I, indoor).

**Table 5**
Average concentrations of IHg in main exposure media.

| Medium | Site A | Site B | THg limit |
|---|---|---|---|
| Indoor air (ng/m$^3$)[a] | 16.9[b] | 20.7[b] | 50 |
| Outdoor air (ng/m$^3$)[a] | 13.6[b] | 13.7[b] | 50 |
| Water (ng/L) | 66.9[c] | 25.6[c] | 1000 |
| Rice (ng/g DW) | 51.6[c] | 26.4[c] | 20 |
| Corn (ng/g DW) | 2.3[d] | 2.3[d] | 20 |
| Fish (ng/g WW) | 233[e] | 233[e] | 500[f] |
| Vegetable (ng/g WW) | 143[g] | 80[g] | 10 |
| Meat (ng/g WW) | 216[c] | 216[c] | 50 |
| Poultry (ng/g WW) | 160[h] | 160[h] | 50 |

[a] Elemental Hg.
[b] This study.
[c] Feng et al., 2008.
[d] Zhang et al., 2010b.
[e] Qiu et al., 2009.
[f] MeHg limit.
[g] Geomean, Feng et al., 2008.
[h] Ji et al., 2006.

related to stockpiles of calcines and mine wastes at site D and E, which was similar with a previous study by Dai et al. (2012).

Dose–response relationship between UHg and the level of air Hg are quantified in occupational settings. Values for air Hg concentration (in μg/m$^3$) are approximately the same as those for UHg concentration (expressed in μg/g Cr) (WHO, 1991). A significant correlation ($r^2 = 0.599$, $p < 0.001$) was found between Hg in air versus urine from various studies at air Hg range of 10 to 50 μg/m$^3$ (Tsuji et al., 2003) and the regression equation is [urine Hg] = 3.24∗[air Hg]$^{0.833}$. Obviously, the simulated UHg concentrations were significantly lower than the measurements in this study, especially at site A, D and E.

For the assessment of Hg exposure through inhalation, we assumed that a person inhaled approximately 20 m$^3$ of air per day (Horvat et al., 2003), which contains 12 m$^3$ indoor air and 8 m$^3$ outdoor air. The daily dietary intake rates were adopted from the official data in the section of "People's Living Conditions" for rural population in Guizhou Statistical Yearbook (GBS, 2012). For calculation, absorption efficiencies of Hg species in diet by human body are considered as approximately 8% for IHg in diet and 80% for Hg vapor (WHO, 1991). The average bodyweight of 60 kg for adult population was according to the second National Physique Monitoring Bulletin (GASC, 2011).

To better understand the pathways of IHg exposure for the local residents, we selected two typical sites in this study: site A, representing site with high IHg exposure (UHg, 12.8 ± 12.4 μg/g Cr);

**Table 6**
Estimated daily intake of IHg through main routes for adults at site A and B and relative contributions.

| Medium | Daily intake | Absorption rate (%) | Site A | | | Site B | | |
|---|---|---|---|---|---|---|---|---|
| | | | PDI without adjustment (μg/kg/d) | PDI (μg/kg/d) | Relative contribution (%) | PDI without adjustment (μg/kg/d) | PDI (μg/kg/d) | Relative contribution (%) |
| Indoor air[a] | 12 $m^3$ | 80 | 0.0034 | 0.0027 | 2.5 | 0.0033 | 0.0041 | 4.7 |
| Outdoor air[a] | 8 $m^3$ | 80 | 0.0018 | 0.0015 | 1.3 | 0.0015 | 0.0018 | 2.1 |
| Water | 2 L | 8 | 0.0022 | 0.0002 | 0.2 | 0.0001 | 0.0009 | 0.1 |
| Rice | 371 g | 8 | 0.3191 | 0.0255 | 23.7 | 0.0131 | 0.1632 | 18.6 |
| Corn | 74.2 g | 8 | 0.0028 | 0.0002 | 0.2 | 0.0002 | 0.0028 | 0.3 |
| Fish | 1.07 g | 8 | 0.0042 | 0.0003 | 0.3 | 0.0003 | 0.0042 | 0.5 |
| Vegetable | 303 g | 8 | 0.7222 | 0.0578 | 53.7 | 0.0323 | 0.4040 | 46.1 |
| Meat | 63.1 g | 8 | 0.2272 | 0.0182 | 16.9 | 0.0182 | 0.2272 | 25.9 |
| Poultry | 5.75 g | 8 | 0.0153 | 0.0012 | 1.1 | 0.0012 | 0.0153 | 1.7 |
| Total | | | 1.30 | 0.11 | | 0.0702 | 0.8236 | |

[a] Elemental Hg.

and site B, representing site with low IHg exposure (UHg, $1.07 \pm 0.45$ μg/g Cr). Average concentrations of IHg in all main exposure media at site A and B were shown in Table 5. Atmospheric Hg data were investigated in this study, and other dietary Hg data were adopted by previous studies which also conducted in this area. The IHg concentrations of water, rice, and vegetable at site A were about 2 times of these at site B, which showed the gradient to the pollution source.

Estimated daily intake of IHg through main routes for adults at site A and B and relative contributions are listed in Table 6. PDI without adjustment were 1.30 and 0.82 μg/kg/d at site A and B, respectively, which exceeded the PTWI for IHg (0.57 μg/kg/d) recommended by JECFA (2010). If taking the absorption efficiencies into consideration, PDIs of IHg at site A and B were 0.11 and 0.07 μg/kg/d, respectively. For the relative contribution from different pathways, consumption of vegetables, rice, and meat, as a whole, accounted for $>90\%$ of the PDI of IHg. Inhalation of indoor and outdoor ambient air accounted less than 7% of the total daily intake. Eating fish, poultry, corn, and drinking water accounted for a small part of the total daily intake ($<2.6\%$). In conclusion, dietary intake, rather than inhalation of Hg vapor, is the main pathway of IHg exposure at both Site A and B with different IHg exposure levels, which is consistent with a previous study (Zhang et al., 2010b). It is reported that MeHg can bio-accumulate in rice seeds in Hg mining areas (Zhang et al., 2010a, Qiu et al., 2008), and therefore the local inhabitants in Wanshan Hg mining area co-exposure to both IHg and MeHg through dietary intake. Co-exposure may elevate risk via additive or synergistic mechanisms and additional research is needed to address this issue. Remediation measures are urgently needed to restore historical Hg polluted soil and to reduce risk of human Hg exposure in Wanshan Hg mining area.

## 4. Conclusions

The average UHg in the study population was 8.32 μg/g Cr, which indicated that the local population is exposed to IHg in a high level. Residents lived within 3 km from the mine waste and calcine heaps showed high UHg concentrations and it demonstrated a gradient of IHg exposure levels with the distance from the pollution source. IHg exposure impairs renal function and SCr can be served as a good biomarker to assess the extent of Hg exposure. Dietary intake, rather than inhalation of Hg vapor, is the main pathway of IHg exposure for the local population in Wanshan Hg mining area.

## Acknowledgments

This study was funded by the National Key Basic Research Program of China (2013CB430004), and the National Natural Science Foundation of China (21007068, 41120134005, and 41373135).

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