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# Human hair mercury levels in the Wanshan mercury mining area, Guizhou Province, China

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**Abstract** The total mercury (T-Hg) and methyl mercury (Me-Hg) concentrations in the hair were measured to evaluate mercury (Hg) exposure for the residents in Da-shui-xi Village (DSX) and Xia-chang-xi Village (XCX) in the Wanshan Hg mining area, Guizhou Province, Southwestern China. The mean concentrations in the hair of DSX residents were 5.5 ± 2.7 μg/g and 1.9 ± 0.9 μg/g for T-Hg and Me-Hg, respectively. The concentrations in the hair of XCX residents were 3.3 ± 1.4 μg/g and 1.2 ± 0.5 μg/g for T-Hg and Me-Hg, respectively. Hair Me-Hg concentrations were significantly correlated to T-Hg ($r = 0.42$, $P < 0.01$) in the two sites; on average, hair Me-Hg concentration accounted for 40 and 44% of T-Hg for DSX and XCX residents, respectively. Age has no obvious correlation with hair Hg and the hair Hg levels showed a significant gender difference, with higher T-Hg and Me-Hg concentrations in the hair from males than females. The rice collected from the two sites showed high levels of T-Hg and Me-Hg concentration. The results indicated a certain Hg exposure for the residents in DSX and XCX in the Wanshan Hg mining area.

**Keywords** Total mercury · Methyl mercury · Hair · Mercury exposure · Wanshan mercury mining area

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## Introduction

Mercury (Hg) is considered as a global pollutant (Lindqvist 1991), due to its long residence time in the atmosphere (from 0.5 to 2 years, Schroeder and Munthe 1998). The long-range transport of atmospheric Hg, its deposition, bioaccumulation, and the enrichment of highly toxic methylmercury (Me-Hg) compounds in the aquatic food chain pose a serious environmental problem, even in remote areas (Lucotte et al. 1995; Schuster et al. 2002; Miller et al. 2005).

Hg can cause significantly adverse effects on human health and the toxicity depends on its chemical forms. The most important chemical forms from a toxicological point view are elemental Hg and Me-Hg. For elemental Hg (Hg vapor), approximately 80% of that which is inhaled is retained the bloodstream and then distributed to the tissues. The elimination occurs via urinary and fecal excretion (WHO 1991). Urinary Hg measurements are widely used for the assessment of exposure to inorganic Hg (mainly Hg vapor) in humans (Barregård 1993).

Inorganic Hg (e.g., Hg vapor) may cause a variety of adverse effects, but the most classic ones are found in the central nervous system (tremor and mental changes) and the kidneys (e.g., proteinuria). The inhalation of Hg vapor for occupational workers and dental amalgamation for the general population are the most important routes of human exposure to Hg vapor (WHO 1991).

Me-Hg is a potent toxicant and its neurotoxicity is well known as a result of the outbreaks of severe poisoning in Japan, Iraq, and elsewhere in the last century. Me-Hg in the diet is almost completely (95%) absorbed into the bloodstream. Hair and blood Hg levels are widely used for the monitoring of Me-Hg exposure (WHO 2000). The toxicity of Me-Hg is much higher than that of inorganic Hg. The nervous system is also the principal target of the effects of Me-Hg in humans. The sensory, visual, and auditory functions, together with those of the cerebellum, which is concerned with coordination, are the most common functions to be affected (WHO 1990). The fetus, newborns, and young children are especially sensitive to Me-Hg exposure because of the sensitivity of the developing nervous system. Me-Hg exposure through fish consumption and its related health effects for humans is a worldwide concern (Mergler et al. 2007).

Mercury contamination from abandoned Hg mines is a great environmental concern around the world. Generally, abundant toxic Hg compounds which are contained in the historical mine waste can be transferred to water, soil, and sediments compartment, and possibly converted into Me-Hg under certain conditions, which eventually enter the human body through food chains (Gray et al. 2002, 2004). Wanshan Hg deposit located in the eastern part of Guizhou Province, southwestern China (Fig. 1), was the largest Hg mine in China. A long history of mining activities have produced serious Hg contamination to the local environment and adjacent ecosystems, including contamination to the air, water, soil, sediments, and organisms (Feng et al. 2003; Horvat et al. 2003; Zhang et al. 2004; Qiu et al. 2005). As far as we know, human Hg exposure surveys are rarely reported in the study area.

The present study was designed to evaluate Hg exposure of the residents in the Wanshan Hg mining area by hair Hg investigation. Previous researches showed that hair total Hg (T-Hg) analysis is difficult in differentiating between exogenous metal contamination and the metal deposited endogenously. Therefore, we measured both T-Hg and Me-Hg concentrations in the hair to distinguish the Me-Hg intake and possible external exposure to Hg vapor. Qiu et al. (2008) demonstrated that rice is an Me-Hg bio-accumulative plant and the main Me-Hg source for human exposure in Hg mining areas was through rice intake (Feng et al. 2008). Thus, rice samples, which is considered as the staple food for the participants, were also collected for Hg analysis and then for daily Hg intake estimation.

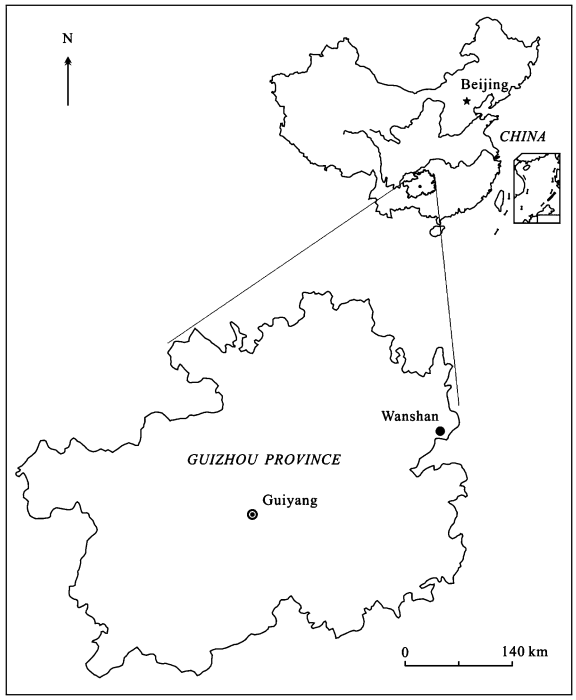


**Fig. 1** Locations of the study areas in Guizhou Province, China

## Materials and methods

### Study area

The mining activities at Wanshan were initiated in the Qin Dynasty (221 B.C.), but completely ceased in 2001. Approximately 22,000 tons of Hg, 6,000 tons of cinnabar, and large quantities of mine waste had been produced at Wanshan (Qiu et al. 2005). Da-shui-xi Village (DSX) and Xia-chang-xi Village (XCX) in the Wanshan Hg mining area were selected for hair

Hg investigation (Fig. 1). DSX is just located at about 1.5 km downstream of Da-shui-xi Stream. Da-shui-xi Dam, which was build to receive drainage and mine waste discharged from the Hg processing facility, is located at the upper end of the stream. And XCX is situated at about 1.5 km downstream of DSX.

### Sample collection

In March 2005, a total of 85 hair samples were collected from the residents of DSX and XCX. Hair samples were cut with 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. Rice samples were collected from each participating family and the rice was cultivated in the local residents' own land. All participants were asked to fill in a questionnaire, including information on age, gender, profession, the history of involvement of artisanal Hg smelting activity, illness, and food consumption, such as average daily intakes of rice, vegetables, meat, and fish.

### Analytical method

Hair samples were washed with nonionic detergent, distilled water, and acetone, and dried in an oven at 60°C overnight. Hair and rice samples were digested in a water bath (95°C) with a fresh mixture acid of $HNO_3/H_2SO_4$ (v/v 4:1) for T-Hg analysis (Horvat et al. 1991). T-Hg concentrations in hair and rice samples were determined by BrCl oxidation, $SnCl_2$ reduction, purge, gold trap, and cold vapor atomic fluorescence spectrometry (CVAFS). For Me-Hg analysis, prepared hair and rice samples were digested using the KOH-methanol/solvent extraction technique (Liang et al. 1994, 1996). Me-Hg contents in hair samples were measured using aqueous ethylation, purge, trap, and GC-CVAFS detection.

### Quality control

The quality control system consists of method blanks, blank spikes, matrix spikes, certified reference material, and blind duplicates. The mean T-Hg concentration obtained from the certified reference material of hair sample (NIES-13) was 4.4 ± 0.1 μg/g ($n = 6$), which accorded well with the certified value of 4.4 ± 0.2 μg/g, and an average Me-Hg content of 3.5 ± 0.1 μg/g ($n = 5$) was found from NIES-13 with the certified value of 3.8 ± 0.4 μg/g. The relative percentage difference was lower than 5% for T-Hg and Me-Hg in hair duplicate samples.

## Results and discussion

### Hair Hg levels

Concentrations of T-Hg and Me-Hg in the hair from DSX and XCX residents are summarized in Table 1. The average value of T-Hg concentration in the hair of residents from DSX was 5.5 ± 2.7 μg/g (mean ± SD), ranging from 1.5 to 16 μg/g, and the average Me-Hg concentration was 1.9 ± 0.85 μg/g (mean ± SD). The mean value of T-Hg concentration in the hair of residents from XCX was found to be 3.3 ± 1.4 μg/g (mean ± SD), varying from 1.6 to 9.4 μg/g, and the average Me-Hg concentration was 1.2 ± 0.48 μg/g (mean ± SD). The mean hair T-Hg and Me-Hg concentrations from DSX and XCX residents were clearly higher than that from Changshun in the Guizhou Province, which was found to be 0.78 ± 0.28 μg/g (mean ± SD) for T-Hg and 0.65 ± 0.25 μg/g for Me-Hg, respectively (Li et al.

**Table 1** Concentrations of T-Hg and Me-Hg in the hair of the studied subjects

| Site | *n* | Age | Sex | | T-Hg (μg $g^{-1}$) | | Me-Hg (μg $g^{-1}$) | | %Hg as Me-Hg | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | M | F | Mean ± SD | Range | Mean ± SD | Range | Mean ± SD | Range |
| DSX | 49 | 49 ± 22 | 28 | 21 | 5.5 ± 2.7** | 1.5–16 | 1.9 ± 0.85* | 0.70–4.4 | 40 ± 19 | 13–93 |
| XCX | 36 | 46 ± 22 | 22 | 14 | 3.3 ± 1.4 | 1.6–9.4 | 1.2 ± 0.48 | 0.71–2.5 | 44 ± 15 | 22–74 |
| Total | 85 | 45 ± 22 | 50 | 35 | 4.6 ± 2.5 | 1.5–16 | 1.7 ± 0.83 | 0.70–4.4 | 41 ± 18 | 13–93 |

* $P < 0.05$ (compared with XCX)

** $P < 0.01$

2008a). The mean hair T-Hg levels also exceeded the normal value (2 μg/g) recommended by the WHO (1990). These observations indicated that the residents in DSX and XCX were exposed to Hg to a certain extent.

Compared with other data from similar studies (Table 2), the hair Hg concentrations in the present study were relatively lower than that of a large amount of a fish-eating population in the Amazon River, which was impacted by amalgamation for gold extraction. And the hair T-Hg concentrations were comparable to the results from the Palawan Hg mine, Philippines (Williams et al. 2000), and the Diwalwal gold amalgam area, Philippines (Drasch et al. 2001).

The mean T-Hg and Me-Hg concentration in the hair samples of DSX residents was significantly higher than that of XCX residents in the Wanshan Hg mining area (T-Hg, $P < 0.01$; Me-Hg, $P < 0.05$), which indicated more serious Hg exposure for the residents in DSX than in XCX. This was in good agreement with the location of the two sites. As mentioned above, DSX is located more closely downstream of the smelting residue heaps than XCX, so that DSX is more seriously contaminated with Hg.

## Correlation between hair T-Hg and Me-Hg

A statistically significant correlation ($r = 0.42$, $P < 0.01$) was observed between T-Hg and Me-Hg concentrations in the hair from DSX and XCX. Me-Hg in the hair accounted for, on average, 40 and 44% of T-Hg for the residents in DSX and XCX, respectively. Most previous studies reported that Me-Hg constituted 70–80% of T-Hg in hair and the major exposure route was through fish consumption (Lebel et al. 1998; Barbosa et al. 2001; McDowell et al. 2004). According to the questionnaires, ten participants from DSX and six participants from XCX were involved or historically involved in artisanal (small-scale) Hg mining activities. A previous study demonstrated that artisanal smelting workers in Guizhou Province were exposed to Hg vapor through inhalation, and, as a result, T-Hg concentrations in the hair and urine were significantly elevated (Li et al. 2008a). Since the population in the

**Table 2** Hair T-Hg concentrations from different exposed populations worldwide

| Location | *n* | Mean ± SD (μg $g^{-1}$) | Range (μg $g^{-1}$) | Comments | References |
|---|---|---|---|---|---|
| Palawan, Philippines | 130 | 3.7 | 0.1–18.5 | Hg mining impacted area | Williams et al. (2000) |
| Ten cities in Japan | 8,665 | 1.82 (GM) | 0.02–29.37 | | Yasutake et al. (2004) |
| Rio Branco, Brazil | 2,318 | 2.418 ± 3.850 | | Urban population | de Oliveira Santos et al. (2002a) |
| Kuwait | 100 | 4.181 ± 3.220 | | Fishermen | Al-Majed and Preston (2000) |
| Tucurui, Para, Brazil | 125 | 35 | 0.9–240 | Fishermen | Leino and Lodenius (1995) |
| Cambodia | 94 | 3.1 (GM) 7.3 (AM) | 0.54–190 | A source other than fish may be responsible for high Hg in some Cambodians | Agusa et al. (2005) |
| Sai Cinza, Para, Brazil | 324 | 16.0 ± 18.92 14.72 (GM) | 4.5–90.4 | Munduruku Indians, fish-eating population | de Oliveira Santos et al. (2002b) |
| Madeira River Basin, Amazon, Brazil | 713 | 15.22 ± 9.60 | 5.99–150 | Riverside population | Bastos et al. (2006) |
| Jacareacanga, Para, Brazil | 205 | 8.6 | 0.3–83.2 | Brazilian Amazon riverine community | Crompton et al. (2002) |
| Diwalwal, Philippines | 316 | 4.14 | 0.03–37.76 | Gold amalgamation area | Drasch et al. (2001) |
| Wujiazhan town on the Di'er Songhua river, northeast China | 108 | 3.44 (AM) 0.648 (GM) | 0.16–199 | The river was polluted with Me-Hg by industrial wastewater discharge | Zhang and Wang (2006) |
| DSX, Wanshan | 49 | 5.5 ± 2.7 | 1.5–16 | Hg mining area | This study |
| XCX, Wanshan | 36 | 3.3 ± 1.4 | 1.6–9.4 | Hg mining area | This study |

study was exposed to Me-Hg mainly through rice digestion (Feng et al. 2008), the low percentages of Hg as Me-Hg in the hair sample might have resulted from the elevation of T-Hg concentration in hair samples through Hg vapor exposure. The rice and vegetables which were collected from the Hg mining area also showed highly elevated inorganic Hg (I-Hg) levels (Horvat et al. 2003; Qiu et al. 2005, 2008; Feng et al. 2008). The mean of I-Hg concentrations was up to 546 ng/g in Gouxi and the percentages of I-Hg as Hg in rice ranged from 7 to 98.6% (Qiu et al. 2008). The percentages of I-Hg as Hg in vegetables from the Wanshan Hg mining area were close to 100% and the mean of I-Hg concentrations varied from 87 to 346 ng/g (Feng et al. 2008). Therefore, the I-Hg exposure through diet ingestion (especially for rice and vegetable) was very high, even though the absorbed proportion of I-Hg is very low (8%; WHO 1991). This may also be responsible for the low percentages of Hg as Me-Hg in the hair samples.

## Age-related variation in hair Hg levels

There are only a few references to age-related differences in the hair Hg levels. Lee et al. (2000) observed that T-Hg and Me-Hg levels tended to increase with the age of subjects in males ($P < 0.01$) for the urban residents of South Korea, but not in females. Yasutake et al. (2004) observed a transient decline around the age of 20s, Hg levels increased into their 50s and 60s, and declined thereafter for the Japanese population. Hormonal control might be involved in Hg uptake by human hair for the Japanese. For the residents in DSX and XCX in the Wanshan Hg mining area, no significant correlations were found between age and T-Hg ($r = 0.05$, $P = 0.68$) or Me-Hg concentrations ($r = 0.01$, $P = 0.94$). The hair Hg concentration for different age groups is given in Table 3.

## Gender difference in hair Hg concentrations

Gender differences in hair Hg levels were found in many reports. Lower levels in women's hair compared to men were generally observed (Shimomura et al. 1980; Airey 1983; Lee et al. 2000). The amount of fish consumed might be responsible for the higher male Hg levels and hormonal control might also be a possible factor (Lee et al. 2000). The Hg levels of hair samples collected from the Wanshan area showed a significant gender difference (T-Hg, $P < 0.05$; Me-Hg, $P < 0.01$), with higher Hg concentrations in the hair from males than females, as shown in Fig. 2.

**Table 3** Hair Hg concentrations in different age groups (μg $g^{-1}$)

| Age | Number | T-Hg | | Me-Hg | |
|---|---|---|---|---|---|
| | | Mean | SD | Mean | SD |
| 0–9 | 6 | 4.0 | 1.3 | 2.6 | 0.7 |
| 10–19 | 11 | 4.6 | 2.0 | 1.4 | 0.5 |
| 21–29 | 4 | 2.8 | 1.0 | 1.4 | 0.8 |
| 30–39 | 8 | 5.0 | 2.8 | 1.7 | 0.3 |
| 40–49 | 12 | 5.0 | 2.4 | 1.6 | 0.7 |
| 50–59 | 19 | 5.0 | 3.4 | 1.7 | 0.7 |
| 60–69 | 18 | 4.2 | 2.1 | 1.6 | 1.0 |
| 70–79 | 5 | 5.4 | 3.7 | 1.5 | 0.4 |
| >80 | 2 | 2.8 | 0.5 | 1.6 | 0.5 |

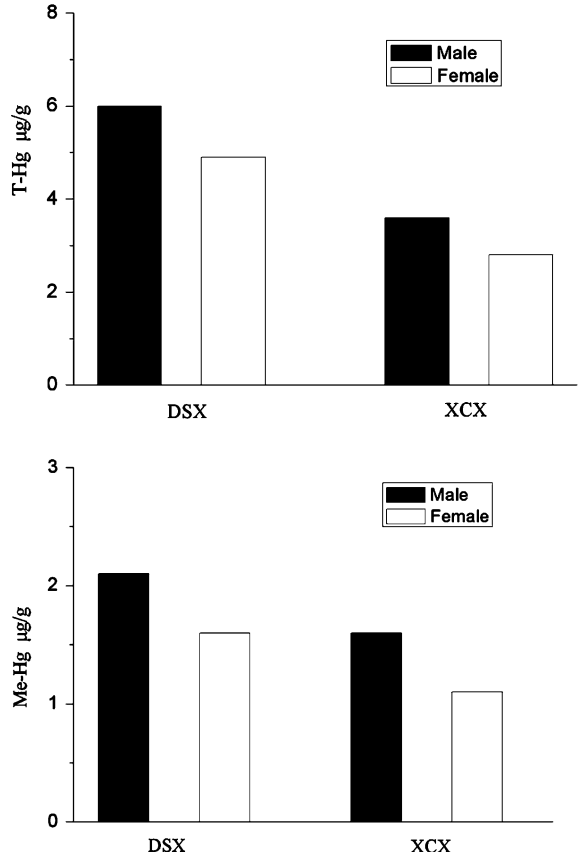


**Fig. 2** Gender differences of hair T-Hg and Me-Hg concentrations

Generally, higher T-Hg concentrations in hair for males might be attributed to occupational exposure of Hg vapor. For Me-Hg, the intake of Me-Hg-contaminated rice was the major route for residents in the Wanshan Hg mining area (Feng et al. 2008). Gender differences on daily rice intake were observed in the population from the Wanshan Hg mining area, which indicated that males consumed a significantly ($P < 0.05$) larger amount of rice than females per day (Feng et al. 2008). Therefore, larger amount of rice consumption was responsible for the higher hair Me-Hg levels for males.

## Rice Hg levels and Me-Hg intake assessment

The concentrations of T-Hg and Me-Hg in the rice samples collected from DSX and XCX are listed in Table 4. The average value of T-Hg concentration in the rice from DSX was found to be 109 ng/g, ranging from 19.0 to 393 ng/g, and that of Me-Hg was up to 10.3 n/g, ranging from 2.51 to 18.1 ng/g. The mean value of T-Hg concentration in the rice from XCX was found to be 23.5 ng/g, ranging from 17.5 to 30.4 ng/g, and that of Me-Hg was up to 7.61 n/g, ranging from 3.47 to 20.9 ng/g. The average T-Hg concentration in rice from the two sites exceeded the maximum concentration of 20 ng/g recommended by Chinese National Standard Agency (1994) to the tolerance limit of Hg in foods.

The mean T-Hg concentration in the rice from DSX was significantly ($P < 0.05$) higher than that from XCX and the mean Me-Hg concentrations in the rice from DSX was higher than that from XCX, but not significantly so ($P = 0.33$). It was in good

**Table 4** Concentrations of T-Hg and Me-Hg in rice samples in the study area

| Sampling site | Sampling number | T-Hg (ng $g^{-1}$) | Me-Hg (ng $g^{-1}$) | %Hg as Me-Hg | Description |
|---|---|---|---|---|---|
| DSX | R1 | 20.6 | 5.16 | 25.1 | Local rice |
| | R2 | 131 | 18.1 | 13.8 | Local rice |
| | R3 | 19.0 | 8.49 | 44.8 | Local rice |
| | R4 | 72.3 | 6.39 | 8.84 | Local rice |
| | R5 | 393 | 17.5 | 4.44 | Local rice |
| | R6 | 84.3 | 7.37 | 8.74 | Local rice |
| | R7 | 125 | 16.9 | 13.6 | Local rice |
| | R8 | 22.6 | 2.51 | 11.1 | Local rice |
| | Total | 109 | 10.3 | 16.3 | |
| XCX | R9 | 20.7 | 8.24 | 39.8 | Local rice |
| | R10 | 21.4 | 3.85 | 18.0 | Local rice |
| | R11 | 17.5 | 3.47 | 19.8 | Local rice |
| | R12 | 25.5 | 8.64 | 33.9 | Local rice |
| | R13 | 29.1 | 6.51 | 22.4 | Local rice |
| | R14 | 30.4 | 4.68 | 15.4 | Local rice |
| | R15 | 18.1 | 4.58 | 25.3 | Local rice |
| | R16 | 25.5 | 20.9 | 82.0 | Local rice |
| | Total | 23.5 | 7.61 | 32.1 | |
| Wanshan market | R17 | 12.5 | 3.91 | 31.3 | Commercial rice from Wanshan market |
| | R18 | 19.8 | 8.71 | 44.1 | Commercial rice from Wanshan market |
| | R19 | 12.9 | 8.37 | 64.7 | Commercial rice from Wanshan market |
| | R20 | 20.1 | 6.39 | 31.8 | Commercial rice from Wanshan market |
| | Total | 16.3 | 6.85 | 43.0 | |
| Guiyang market | R21 | 3.36 | 1.99 | 59.3 | Commercial rice from Guiyang market |
| | R22 | 2.15 | 0.62 | 28.8 | Commercial rice from Guiyang market |
| | Total | 2.76 | 1.31 | 44.0 | |

**Table 5** Comparison of Hg concentrations in rice collected from different areas

| Location | T-Hg (ng $g^{-1}$) | Me-Hg (ng $g^{-1}$) | %Hg as Me-Hg | Authors |
|---|---|---|---|---|
| Wanshan Hg mining area, China | 11.1–569 | 8.03–144 | 5.46–72.6 | Horvat et al. (2003) |
| Qingzhen Hg polluted area, China | 2.53–33.5 | 0.71–28.0 | 28.1–83.7 | Horvat et al. (2003) |
| Wuchuan Hg mining area, China | 8.8–550 | 1.2–18 | 2–66 | Qiu et al. (2006) |
| Wuchuan Hg mining area, China | 6.0–113.0 | 3.1–13.4 | 6.0–83.6 | Li et al. (2008b) |
| Wanshan Hg mining area, China | 10.3–1120 | 1.61–174 | 1.4–93 | Qiu et al. (2008) |
| Wanshan Hg mining area, China | 17.5–393 | 2.51–20.9 | 4.44–82.0 | This study |

**Table 6** Estimated daily Me-Hg intake by rice for residents in DSX and XCX

| Site | Rice Me-Hg (ng $g^{-1}$) | | Daily rice intake (g $d^{-1}$) | Body weight (kg) | Daily Me-Hg intake (μg $d^{-1}$ $kg^{-1}$) | |
|---|---|---|---|---|---|---|
| | Mean ± SD | Range | | | Mean ± SD | Range |
| DSX | 10.3 ± 6.2 | 2.51–18.1 | 600 | 60 | 0.103 ± 0.062 | 0.025–0.181 |
| XCX | 7.61 ± 5.7 | 3.47–20.9 | 600 | 60 | 0.076 ± 0.057 | 0.035–0.209 |

agreement with the general observation that the concentrations of T-Hg and Me-Hg decreased with distance from the contamination source. Because DSX is located upstream, the rice was more seriously polluted with Hg. The mean T-Hg and Me-Hg concentrations in the rice from the two studied sites were much higher than that from Wanshan market and Guiyang market. The results were comparable to that which other studies obtained in rice samples in Hg-polluted areas (Horvat et al. 2003; Qiu et al. 2006, 2008; Li et al. 2008b) and the comparisons are presented in Table 5. The results indicated that rice produced in different Hg mining areas in Guizhou Province show high accumulation abilities of Me-Hg. Feng et al. (2008) confirmed that the main route of human Me-Hg exposure is through frequent rice meals intake, but not via fish consumption in Hg mining areas in Guizhou Province, China.

In this study, we assumed that the daily consumption of rice is 600 g and there is no Me-Hg lost during cooking (Horvat et al. 2003). Estimated daily Me-Hg intakes are presented in Table 6 and are expressed as μg $kg^{-1}$ on a body weight basis of a 60-kg person. The daily Me-Hg intake did not exceed 0.23 μg $kg^{-1}$ $d^{-1}$, which is the value recommended by the WHO (2003). In 1997, the US Environmental Protection Agency set a new guideline for Me-Hg in the diet of 0.1 μg $kg^{-1}$ $day^{-1}$ (USEPA 1997). However, the intakes in some populations of the study areas exceeded the new USEPA recommended values. Me-Hg intake through rice consumption, therefore, present a health risk for the residents in DSX and XCX in the Wanshan Hg mining area.

## Conclusion

Hair Hg levels indicated that the residents in Da-shui-xi Village (DSX) and Xia-chang-xi Village (XCX) in the Wanshan Hg mining area were exposed to Hg in a certain content. Age has no obvious correlation with hair Hg and gender differences were found which indicated that males manifested higher concentrations than females for both hair T-Hg and Me-Hg concentrations. Rice with a high concentration of Me-Hg can pose a threat of Me-Hg exposure to the local residents.

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