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1 **The impact of an abandoned mercury mine on the environment in**  
2 **the Xiushan region, Chongqing, southwestern China**

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25

26 **Abstract**

27 Mercury (Hg) contamination is a serious problem at Hg mining area of Xiushan  
28 County, Chongqing, southwestern China. Concentrations of total mercury (THg) and  
29 methylmercury (MeHg) in paddy soil, surface water, and rice (grain) samples  
30 were analyzed to exhibit regional distribution of Hg contamination. Simultaneously  
31 gaseous elemental Hg (GEM) in ambient air in the vicinity of sampling sites were  
32 measured in situ. Hg concentrations in surface water were highly elevated ranging  
33 from 13-2390 ng/L, and MeHg concentrations varied between 0.17-1.1 ng/L.  
34 Dissolved Hg and MeHg concentrations in surface water ranged from 4.7-470 ng/L  
35 and 0.14-0.35 ng/L, respectively. High THg and MeHg concentrations were also  
36 obtained in paddy soils from mining areas, ranging from 0.45-68 µg/g and 0.13-4.8  
37 ng/g, respectively. Similar to high concentrations in water and soil, Hg in rice (grain)  
38 ranged from 4.7-550 ng/g and MeHg from 2.9-26 ng/g. Elevated Hg concentrations in  
39 rice, as a staple food to residents, confirmed that rice consumption could be a vital  
40 pathway of MeHg exposure to native populations. The spatial distribution  
41 characteristics of Hg and MeHg pollution in the local environment indicated the  
42 origin from historic Hg mining sites in the Xiushan area.

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44

45 **Keywords:** Mercury; Methylmercury; Paddy soil; Surface water; Rice; Hg mining

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48 **1. Introduction**

49 Mercury (Hg), especially methylmercury (MeHg), is highly toxic which has  
50 raised widely environmental concerns (Clarkson, 1998; UNEP, 2002). Hg can exist in  
51 the atmosphere as gaseous elemental Hg (GEM), and can be migrated for a long  
52 distances due to its long lifetime (0.5-2year) in the atmosphere (Schroeder et al.,  
53 1998). Both inorganic and organic forms exist in the environment, and inorganic  
54 mercury (IHg) can be transformed into MeHg, the most toxic Hg form. The health  
55 risks posed by MeHg due to its bio-magnification in the aquatic ecosystem are well  
56 known. Trace amounts of MeHg in water can lead to much higher and harmful  
57 concentrations of MeHg in fish and their predators. The consumption of fish has been  
58 considered as the major pathway for human MeHg exposure (WHO, 1990).

59 Mercury contamination is a serious problem in China, especially in historical Hg  
60 mining regions in China (Horvat et al., 2003; Qiu et al., 2005; 2006a, b; 2008; 2012;  
61 Feng et al., 2008). The major concern about Hg is its bio-accumulation, the  
62 transformation from IHg to MeHg. Such strongly toxic Hg substances are readily  
63 migrated to environmental compartments of water, soil and sediments. Generally,  
64 retorted ores (termed calcines) originated by low efficiency roasting of cinnabars in  
65 Hg mining areas contain plenty of water soluble Hg compounds, such as the elemental  
66 Hg and Hg salts (Kim et al., 2000; 2004). Under certain conditions, those soluble Hg  
67 compounds can be transformed into MeHg and finally flow into the human body  
68 through the food chain (Watras et al., 1992; Weber, 1993). Though localized  
69 ecological degeneration and health exposure risk from these contaminated sites, their  
70 combined impacts arise more widespread concern. Usually, the local residents of the  
71 mining districts are exposed to long term health problems (Bose-O'Reilly et al.,  
72 2010).

73 The Xiushan Hg mine, namely Yangshikeng (YSK), is situated in the  
74 southeastern part of Chongqing, southwestern China. The Hg mining activity of state-  
75 owned company at the YSK started at 1960s with a capacity of 25 tons metallic Hg  
76 per year. Currently a private company of Yameiou holders the mining permit of the  
77 YSK and its yearly output of Hg is approximately 30 tons. During sampling

78 campaigns a few of illegal artisanal Hg mining sites in the region were also observed.  
79 Rice was the staple food for native residents.

80 Currently, rice was proved to have strong enrichment ability of MeHg and rice  
81 consumption was confirmed to be the major MeHg exposure pathway for local  
82 residents in Hg mining area (Feng et al., 2007; Qiu et al., 2008; Li et al., 2010).  
83 However, up to now, only some THg mercury data was reported (Li, 2014; Zhang et  
84 al., 2011). Due to the smaller scale than Wanshan, and Xunyang, few attention has  
85 been paid in this area. There is still lack of research conducted to reveal the MeHg  
86 contamination in environmental media for further human health risk assessment in  
87 Xiushan Hg mining area.

88 In this study, we analyzed THg and MeHg concentrations in water, rice and paddy  
89 soil. Gaseous elemental mercury (GEM) in the ambient air were also measured in situ.  
90 The purposes of this study are to evaluate the Hg pollution status in Xiushan by  
91 examining (1) GEM in the atmosphere; (2) Hg speciation in surface water as well as  
92 THg and MeHg in rice and soil; and (3) potential environmental risks from MeHg  
93 containing rice to local residents. The research is latest one in a range of researches  
94 related to abandoned Hg mining area in China in our research team, including several  
95 published studies of the Wanshan (Qiu et al., 2005), Wuchuan (Qiu et al., 2006a),  
96 Lanmuchang (Qiu et al., 2006b), Tongren (Li et al., 2008a), Xunyang (Qiu et al.,  
97 2011, 2012), and Yanwuping Hg mine (Qiu et al., 2013).

## 98 **2. Study area**

99 The Yangshikeng (YSK) Hg mining district (E: 108°53'2"-108°54'1"; N:  
100 28°34'5"-28°36'4"), is located 32 km northwestern of the Xiushan County, covering  
101 approximately 2.5 km<sup>2</sup> (**Figure 1**). The district is in a karstic and hilly region with  
102 elevation ranging from 1631 to 246 m. It possesses a subtropical humid climate  
103 featured by mild temperature and rich precipitation. The annual mean temperature is  
104 16 °C. Average annual rainfall is about 1341 mm. Cinnabar is the main mineral in the  
105 YSK is with grade ranging from 0.13% to 0.28%. The Shileixi creek is an affluent of  
106 the Rongxi River, which is directly affected by the Hg mining activity in history and  
107 runs through the YSK area. And there exist 18.3 thousand residents living in the

108 downstream of YSK area, most of them are farmers who depend on the yield from the  
109 rice paddy fields. Most of mine-wastes were deposited along the Shileixi Creek  
110 without control. Significant quantities of mine-waste calcines were heaped around  
111 abandoned mining and retorts processing sites.

112

### 113 **3. Methods**

#### 114 **3.1. Samplings and preparation**

115 The sampling of soil, water, and rice was carried out in September 2011. During  
116 the sampling time, it was clear and the river runoff was at base flow condition. Water  
117 samples (both unfiltered and filtered surface water, n=11) were collected from the  
118 Shileixi creek river for Hg and MeHg analysis. Among samples, X15 is a sample of  
119 leachate from abandoned mine-waste tailings of the YSK. Unfiltered samples were  
120 directly gathered from the river for THg and total MeHg (TMeHg). Meanwhile, ultra  
121 cleaned syringes and membrane filters (0.45  $\mu\text{m}$ ) were used for filtered samples for  
122 Dissolved THg (DHg) and Dissolved MeHg (DMeHg). Ultra-pure HCl were added  
123 into water samples on-site, obtaining an acid concentration of 0.4% (v/v) according to  
124 Method 1631E ([USEPA, 2001](#)). All the water samples were collected and preserved in  
125 borosilicate glass bottles pre-cleaned by heating in a muffle furnace at 550° C for 3 h.

126 Soil samples (n=15) were gathered from paddy fields within or nearby the  
127 Shileixi creek and Rongxi river (**Figure 1**). Each soil sample is a composite sample of  
128 3-5 sub-samples from an area of 1 m<sup>2</sup> in diagonal sampling technique. Rice paddy soil  
129 samples for all geochemical analysis were sealed in plastic bags to prevent cross  
130 contamination. In the laboratory, all samples were kept at -20 °C in a refrigerator, then  
131 appropriate amounts of samples were freeze-dried, homogenized in an agate mortar,  
132 and passed through minus 200-mesh sieve.

133 Rice grain samples (n=15) were collected at the same time from to the same  
134 locations of soil samples. All the rice samples were sealed and stored in the same way  
135 as soil samples to prevent cross contaminations. Then the rice samples were freeze-  
136 dried, and ground with grinder (IKA-A11), and then sieved through 200 meshes, and  
137 stored in plastic bags for analysis.

138 Ambient air Hg (GEM, n=22) concentrations were measured in situ within the  
139 vicinity of the soil sampling locations. The Lumex RA-915<sup>+</sup> (Lumex Ltd., Russia)  
140 was employed to measure GEM, and the detection limit for atmospheric Hg is 0.5  
141 ng/m<sup>3</sup> at the rate of 10 L/min. For the machine, after 5 min for obtaining stable  
142 baseline stage, an observation lasts for 30-60 min was conducted at each site.

### 143 **3.2. Analytical methods**

144 For rice THg analysis, approximately 0.5-1.0 g of rice sample was dissolved at  
145 95-140 °C with a mixture of HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> (4:1, v/v) (Qiu et al., 2012; 2013). Rice  
146 samples were measured using cold vapor atomic fluorescence spectroscopy (CVAFS)  
147 in accordance with Method 1631E (USEPA, 2002). For rice MeHg determination,  
148 approximately 0.2-0.5 g rice sample was weighed for digestion using 25% KOH-  
149 methanol at 75-80 °C for 3 h. Then MeHg in rice samples was leached with  
150 dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>) and back-extracted into water phase for determination  
151 based on Method 1630 (USEPA, 2001; Liang et al., 1996).

152 For soil THg measurement, about 0.1-0.3 g of sample was heated at 95 °C for 30  
153 min using water bath in a mixed solution of HCl and HNO<sub>3</sub> (3:1, v/v), then BrCl was  
154 added and the mixture heated to 95 °C for another 30 min (Feng et al., 2013; Meng et  
155 al., 2016). Then an appropriate amount of digestive soil solution was determined  
156 using cold-vapor atomic absorption spectrometry (CVAAS) (Feng et al., 2007; Qiu et  
157 al., 2008). For soil MeHg determination, about 0.2-0.5 g soil sample was extracted  
158 with CH<sub>2</sub>Cl<sub>2</sub> after HNO<sub>3</sub> leaching (Liang et al., 1996). Then a proper amount of  
159 digestive solution was used for determination by GC-CVAFS according to Method  
160 1630 (USEPA, 2001).

161 For water THg and DHg, water samples were determined by BrCl oxidation and  
162 SnCl<sub>2</sub> reduction (Bloom et al., 1983). Then appropriate amount of solution was  
163 measured by CVAAS. For TMeHg and DMeHg, water samples were distilled, NaBEt<sub>4</sub>  
164 ethylated, and analyzed by cold vapor atomic fluorescence spectrometry (CVAFS)  
165 (Horvat et al., 1993; Liang et al., 1994, 1996). Specifically, a 45 mL aliquot sample  
166 was added in a Teflon distillation vessel on an Al heating pan and distilled at 170 °C  
167 for 3-4 h. The distillate then proceeded aqueous phase ethylation by adding 0.2 mL 2

168 M sodium acetate and 0.06 mL 1% sodium tetraethylborate step by step, then purging  
169 with N<sub>2</sub> to absorb MeHg onto a Tenax trap (Guo et al., 2008). MeHg was ultimately  
170 desorbed with heating onto an isothermal GC column for peak separation and  
171 analyzed by CVAFS (Brooks Rand Model III).

172 The pH of soil was measured by taking 10 g from samples into 25 ml of reagent  
173 water (Chinese National Standard Agency, 1988). For soil organic matter, the  
174 K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>/ H<sub>2</sub>SO<sub>4</sub> method was used to digest soil samples and then determined by  
175 spectrophotometer according to the national environmental protection standard of  
176 China. For Humic acid, Fulvic acid, and Humin, Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub>, NaOH, K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, and H<sub>2</sub>SO<sub>4</sub>  
177 were used according to the method recommended by the Ministry of Agriculture of  
178 China (2010).

### 179 3.3 QA/QC

180 QA and QC of THg and MeHg analysis were conducted using duplicates, method  
181 blanks, matrix spikes, and certified reference materials (GBW07405; BCR580, TORT-  
182 2).

183 An average total Hg concentration of 0.30±0.01 µg/g (n=5) for the soil standard  
184 (GBW07405) was obtained, which was in accordance with certified value of  
185 0.29±0.04 µg/g. For soil MeHg, BCR 580 was determined and the achieved MeHg  
186 concentration of 74±5 ng/g (n=5) confirm the certified value of 75±4 ng/g. In soil  
187 duplicates, the relative percent difference was less than 10% for THg and 8.4% for  
188 MeHg. The detection limits for THg and MeHg were 0.01 ng/g and 0.002 ng/g,  
189 respectively.

190 In rice, recovery rates on matrix spikes were 91-109% for Hg and 87-105% for  
191 MeHg. TORT-2 was determined, and the obtained value (150±8 ng/g, n=5) met the  
192 certified value (152±13ng/g). In rice duplicates, the relative percentage difference was  
193 less than 7% for Hg and 8.6% for MeHg. The detectable limits for Hg and MeHg  
194 were 0.015 ng/g and 0.004 ng/g, respectively.

195 In water, recovery rate of matrix spikes were 92-109% for THg and 89-112% for  
196 MeHg. The determination limits of THg and MeHg were 0.02 ng/L and 0.009 ng/L,  
197 respectively. The relative percentage difference was <9 % for THg and <8.8% for



198 MeHg in water duplicates. The method blank cannot be detected. The field blank was  
199  $0.15 \pm 0.04$  ng/L for THg and 0.014 ng/L for MeHg, respectively.

200 For soil organic matter, the  $K_2Cr_2O_7/H_2SO_4$  method was used to digest soil  
201 samples and then determined by spectrophotometer according to the national  
202 environmental protection standard of China. For Humic acid, Fulvic acid, and Humin,  
203  $Na_4P_2O_7$ , NaOH,  $K_2Cr_2O_7$ , and  $H_2SO_4$  were used according to the method recommended  
204 by the Ministry of Agriculture of China (2010)

## 205 **4. Results and discussion**

### 206 **4.1. Water**

207 The THg concentrations of unfiltered surface waters from the YSK mining region  
208 ranged from 13 to 1004 ng/L and averaged  $180 \pm 310$  ng/L. However, the leachate from  
209 nearby abandoned large mine-waste tailings (calcines), sample Y13, exhibited  
210 extremely high concentration of 2390 ng/L (**Table 1**). The THg concentrations  
211 decreased to 150 ng/L at 4 km downstream of the tailing. The highest Hg  
212 concentration obtained in leachate draining from the huge calcines pile is as high as  
213 2390 ng/L. The DHg concentrations of filtered waters altered from 4.7 to 470 ng/L  
214 with the average of 68 ng/L. Similarly, the DHg concentrations were also variable and  
215 the highest value was also found in sample Y13. The observations of high THg and  
216 DHg in leachate are in accordance with previous studies on other Hg mining regions  
217 in China (Qiu et al., 2005; 2006a, b; 2009; 2012). The source of Hg released into the  
218 creek is mainly attributed to the waste calcines, the waste calcines contains large pool  
219 of labile and water-soluble Hg which is main contributor for DHg concentration in the  
220 creek (Gray et al., 2002, 2003; Kim et al., 2000, 2004).

221 The concentration of THg in filtered waters and unfiltered waters showed a  
222 sharply decrease with the downstream distances increase (**Figure 2**). At the upstreams  
223 of the rivers, the THg concentrations were quite low compared to the value of 50 ng/L  
224 (Type I, water for headwater and natural reserves, Chinese EPA, 2002) set by Chinese  
225 Hg standard for surface water. However, two samples of Y13 and Y14 exceeded the  
226 1000 ng/L Chinese drinking water Hg guideline (GB5749-2006) and the 770 ng/L  
227 U.S. Environmental Protection Agency (USEPA, 1992) guideline recommended to

228 protect against chronic effects to aquatic wildlife. The high level of THg in water  
229 could therefore pose risks for both human and animals depending on the water as their  
230 drinking sources.

231 The TMeHg and DMeHg of were relatively similar, ranging from 0.17 to 0.55  
232 ng/L, and from 0.14 to 0.35 ng/L, with the mean concentrations of 0.42 ng/L and 0.23  
233 ng/L, respectively. Surprisingly, the highest TMeHg of 1.1ng/L was obtained in  
234 sample Y15, which is not the nearest sample in the downstream of YSK,  
235 demonstrating there may exist favorable conditions for Hg methylation. Meanwhile,  
236 the DMeHg rate (DMeHg/TMeHg) varied from 0.16 to 0.79 with the mean value of  
237 0.63, and the lowest DMeHg rate was 0.16 obtained in Y15, suggesting the most of  
238 MeHg in Y15 may bond to the particles (Qiu et al., 2011).

The THg concentrations of surface waters, gathered from the YSK Hg mining region, were comparable to those obtained in Hg mining regions worldwide (**Table 2**). Waters, influenced by calcines in Coast Range Hg mines, California, and in Palawan Quicksilver Hg mine, Philippines, exhibited the concentrations of 45000 ng/L and 31000 ng/L in Hg, respectively (Rytuba, 2000; Gray et al., 2003). The highest concentration of THg in surface water was reported to reach 23500 ng/L in China Hg mining regions (Zhang et al., 2009). Horvat et al. (2003) and Qiu et al. (2005) also reported that THg in the surface water ranged from 3.3 to 10580 ng/L, with highest concentrations emerged near the mine-waste tailings in the Wanshan Hg mining region. Similarly, the creek waters influenced by the leachate through calcines demonstrated high levels of Hg, which confirm that the mine-waste calcines is the mostly substantial Hg releasing source. The elevated Hg concentrations in waters, especially the two sites (site Y13 and Y14) beyond the Chinese drinking water Hg guideline (1000 ng/L) and chronic effects to wildlife in aquatic systems (770 ng/L) issued by USEPA (1992), which has adverse impacts on the growth and development of aquatic animal.

Moreover, in this study, the TMeHg concentrations is similar to that of water samples from Hg mine in Alaska, Idrija, Palwan, Yanwuping, Xunyang (**Table 2**). However, the MeHg concentrations are less than the results obtained in California (47 ng/L),,

Wanshan (25 ng/L), Almadén (30 ng/L) (**Table 2**), this indicated that the local conditions in Xiushan is less favorable for Hg methylation (Rytuba 2000; Gray et al., 2004; Qiu et al., 2005). Meanwhile, the elevated MeHg may also result in Hg accumulation in wildlife and the aquatic system, which will pose a serious threat to human health (Wan et al., 2004).

#### 4.2. Soil

In the present study, THg concentrations in paddy soils varied from 0.45 to 68  $\mu\text{g/g}$  with a mean of  $9.8 \pm 17.5 \mu\text{g/g}$  (**Table 3**). The sample Y11, affected by mine-wastes discharged from an abandoned artisanal retort along the bank of Shileixi creek, manifested the highest concentration (**Figure 3**). Elevated THg were also obtained in the samples from the sites adjacent to abandoned Hg mines, such as Y10 and Y17, containing 13  $\mu\text{g/g}$  and 15  $\mu\text{g/g}$ , respectively.

Paddy soils from the YSK mining district also showed high MeHg concentrations, ranging from 0.21-3.7 ng/g with the mean value of 1.36 ng/g. In the upstream, soil MeHg concentrations were generally low (<1.1 ng/g). MeHg concentrations were higher in the samples which were closer in distance to the abandoned mine tailings, and the peak value (3.7 ng/g) was observed in artisanal Hg mines (Y11). In the downstream, soil MeHg decreased with the distances away from artisanal Hg mines.

In this study, soil THg exhibited a typical distribution that downward trends appeared with the distance farther away from artisanal Hg mines both in upstream and downstream. The surface water runs through the mining sites, which were seriously polluted by Hg (Qiu et al., 2012; 2013), rice paddies are therefore affected by the incoming water when paddy fields are drowned during the rice growing season. Water from upstream will bring down Hg-enriched particles transported from upstream-contaminated sites during flooded seasons (Horvat et al., 2003). Soil MeHg had the similar distribution tendency to soil THg. Hg contaminated mine-water runoffs were frequently applied to irrigate paddy fields near Hg mining and retorting sites and periodic irrigation in the paddy fields can produce anaerobic conditions in flooded soil

(Gilmour and Henry, 1991; Weber, 1993; Horvat et al., 2003). Under the appropriate pH, organic matter, anaerobic condition, sulfate reducing bacteria and other Hg methylation bacteria will play a key role to convert Hg into MeHg (Qiu et al., 2008; 2013; Rothenberg et al., 2014)

Mineral residue and calcines in the YSK Hg mining area are the major sources of Hg pollution for the local environment. For comparisons, concentrations of THg and MeHg in soils from other Hg mining areas are present in **Table 3**. In comparison with the results in this study, at Almadén, Yanwuping, Idrija, and in California, Alaska, Palawan, Wanshan, Xunyang, higher THg in soils were obtained. Similar to the results for high THg, MeHg concentrations of 7.3 ng/g, 11 ng/g, 23 ng/g, 41 ng/g, and 80 ng/g in soil from the Yangwuping, Xunyang, Wanshan, Alaska, and Idrija mining area were higher than the highest concentration obtained in this study. Though THg and MeHg concentrations in soil samples from the YSK mining area (up to 68 µg/g THg, and 3.7 ng/g MeHg) are less than those obtained from worldwide Hg mines, soils in the YSK mining district are still heavily polluted with Hg compared to 1.0 µg/g regulated by Chinese standards quality of soil (GB15618, 2008). The remarkable concentration present in the surface soils are an obvious index of the Hg mining activities.

### 4.3 GEM

The GEM concentrations varied from 29 to 4209 ng/m<sup>3</sup>, with an average of 498 ± 682 ng/m<sup>3</sup>. The spatial distribution was featured by the remarkable variation and some extremely elevated concentration (**Figure 4**). The highest concentration was obtained at the downwind direction site (Y11) of the cinnabar ores retorting facility, indicating that the retorting activity and its nearby huge pile of mine-waste tailings may be an origin of Hg<sup>0</sup> in the ambient air.

In the current study, the GEM concentrations were remarkably high by comparison with the results reported in urban cities and remote regions in China (Fu et al., 2012). Compared to other historic Hg mining regions in China, which were in the range of 7.4-410 ng/m<sup>3</sup>, and 12-180 ng/m<sup>3</sup>, in Xunyang, and Wanshan (Qiu et al.,

2012, 2013), the average GEM value ( $498 \pm 682 \text{ ng/m}^3$ ) achieved in this study was significantly elevated. The average value of GEM in the YSK Hg mining area is about ten folds of the air quality reference standard for type II ( $50 \text{ ng/m}^3$ ) issued by the Ministry of Environment Protection of China (MEP), posing a potential risk to local residents.

#### 4.4. Rice

Results of rice samples and corresponding locations were shown in **Table 2** and **Figure 1**. Samples gathered within and near Hg mining sites contained elevated concentration of THg, varying from 12 ng/g to 384 ng/g, with the mean value of  $47.5 \pm 97.3 \text{ ng/g}$ . MeHg concentration in rice are also high, varying from 9.2 ng/g to 64 ng/g with the mean value of  $12.1 \pm 15 \text{ ng/g}$ . The peak concentrations of THg and MeHg were 384 ng/g, 64 ng/g collected from abandoned artisanal Hg mines. The rice collected far away from Hg mining sites, exhibited generally low Hg, ranging from 4.7-18 ng/g.

There exists remarkable positive relationships between rice MeHg ( $\text{MeHg}_R$ ), and soil THg ( $r=0.975$ ,  $p<0.01$ ) (**Table 4**). Significant positive correlations can also be observed between GEM and soil THg ( $r=0.995$ ,  $p<0.01$ ), which implying that soil THg may be a source to GEM, and the freshly Hg deposition in soil is readily to be methylated (Meng et al., 2010). The positive relationship ( $r=0.999$ ,  $p<0.01$ ) between rice THg and soil THg indicates that soil THg can contribute a lot to rice THg (Yin et al., 2013).

In comparison with rice samples from unpolluted region, which mean Hg of  $3.3 \pm 1.3 \text{ ng/g}$  and MeHg of  $2.9 \pm 1.0 \text{ ng/g}$  (Meng et al., 2010), samples gathered from the YSK Hg mining district contained high concentrations of both THg and MeHg. For rice samples in the present study, peak concentration of 384 ng/g and 120 ng/g in THg were 19 times and 6 times of the Hg limit (20 ng/g) suggested by the National Food Safety Standard (GB 2672, 2012). Meanwhile, there also existed other sites downstream from Hg mines are higher than the limit. This phenomenon displayed that rice plants in the area were seriously polluted with Hg, posing a potential risk and harm of Hg to the local residents.

The probable daily intake (PDI) values of IHg and MeHg for general rice consumption adult were estimated. It was supposed that regular amount of rice intake was 400 g per day, meanwhile body weight for one person was 60 kg. The evaluated PDI ranged widely from 0.031 to 2.56 mg/kg (mean: 0.32 mg/kg) for THg and from 0.061 to 0.427 mg/kg (average: 0.081 mg/kg) for MeHg. In our search, this large range demonstrates the wide range of Hg concentrations obtained in rice. The PDI value of 0.427 mg/kg for MeHg might suggest that rice consumption is an important approach for MeHg entering into human body, which poses a risk to human health. PDI values of IHg at abandoned Hg mine sites were 0.8 and 2.56 mg/kg per day, which were higher than the PTWI for IHg (0.57 mg/kg/d) issued by JECFA (2010). For MeHg, the highest PDI value of 0.427 mg/kg is more elevated than the new dietary reference dose (RfD) of 0.23 mg/kg body weight per day certified by the UN Committee (WHO, 2003), however the second PDI value of 0.14 mg/kg is slightly higher than the RfD of 0.1 mg/kg body weight per day recommended by the USEPA (1997).

#### **4.5. Correlation between MeHg and organic matter**

Notable positive correlations ( $r=0.788$ ,  $p<0.01$ ) was found between soil MeHg and humic acid (Table 5). Meanwhile, significant positive relationships ( $r=0.605$ ,  $p<0.05$ ) can be observed between soil MeHg and Fulvic acid. These positive relationships implied that humic acid may contribute a lot to affect the MeHg concentration in soil, and fulvic acid can also influence the MeHg in soil in some degree (Hintelmann et al., 1995, 1997). MeHg in rice tissue exhibited a decreasing trend, when increase the humic acid in hydroponic experiment (Su et al., 2016). The relationship in this study could be an explanation for the low MeHg in rice tissues, and humic acid may be bond to the MeHg and thus decrease the mobility of MeHg to the aboveground of rice plant (Ullrich et al., 2001).

## **5. Conclusions**

Elevated concentrations of THg and MeHg in both filtered and unfiltered surface

water can be primarily attributed to the calcines from retorting ores. High concentrations of THg and MeHg in paddy soils in the vicinity of the abandoned artisanal Hg mines were observed. GEM levels in the atmosphere around demonstrated similar distribution pattern to the soil Hg concentration. Rice MeHg had a positive correlation with soil MeHg, indicating soil MeHg may be the major source of MeHg in rice. The significantly positive correlations between soil MeHg and humic acid indicated its key role in controlling the MeHg production in soil. Results of this study point out that the artisanal Hg mines and abandoned calcines tailings caused seriously Hg pollution in Xiushan County, and the concentration levels of Hg in both water and rice exceed the safe level and therefore pose potential health risk for local residents.

### **Acknowledgments**

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## Figures

**Figure 1** The locations of sampling points in Xiushan area, China.

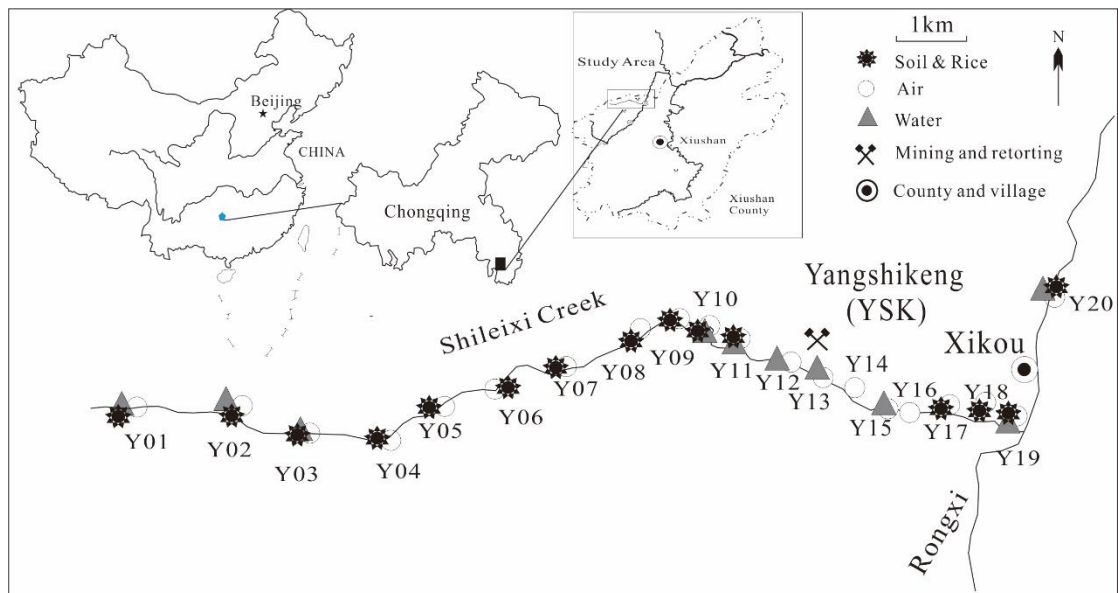
**Figure 2** The distribution of different Hg species in water samples from the YSK Hg mining area, Chongqing, China.

**Figure 3** The THg and MeHg distribution in paddy soils gathered from the YSK Hg mining area in Xiushan County, Chongqing, China.

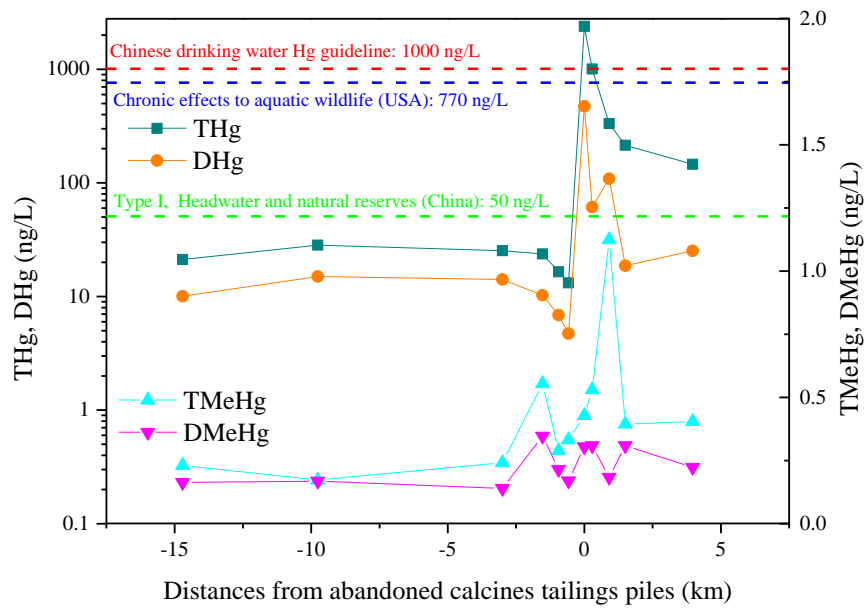
**Figure 4** The GEM concentrations distribution in the ambient air influenced by artisanal Hg mines in Xiushan Hg mining region.

**Figure 5** The Correlations between soil MeHg and humilic acid, and fulvic acid in our research.

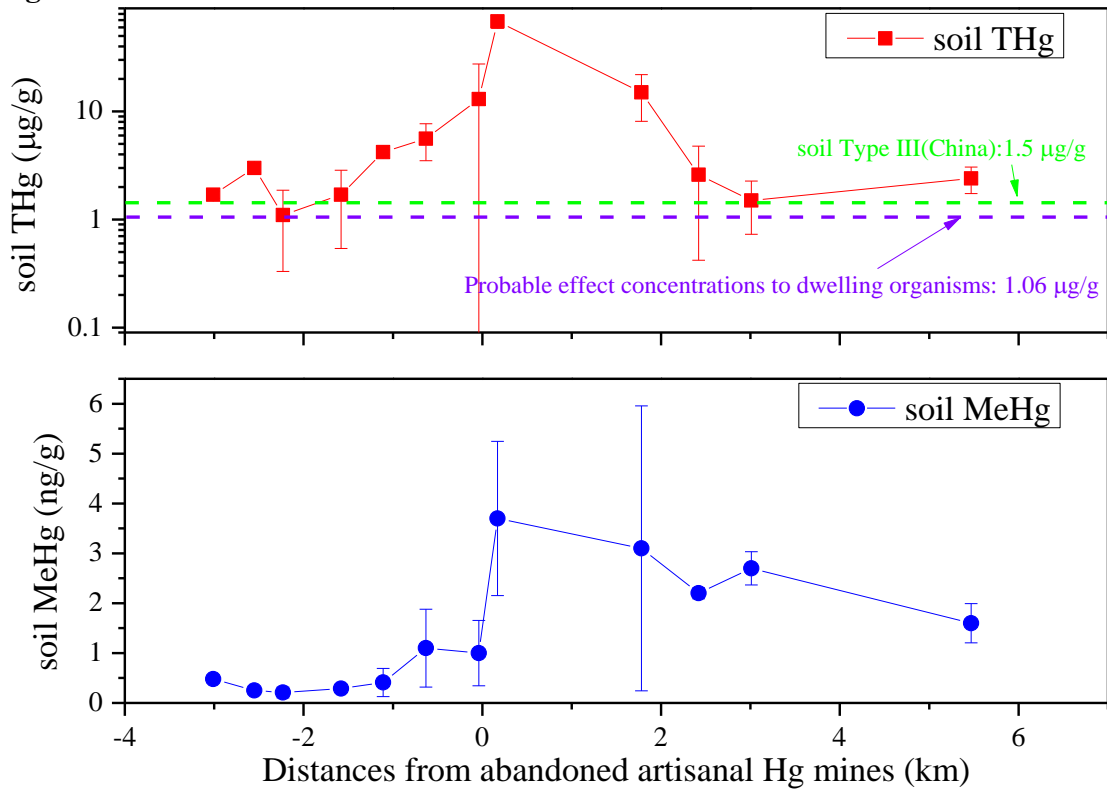
**Figure 1**



**Figure 2**

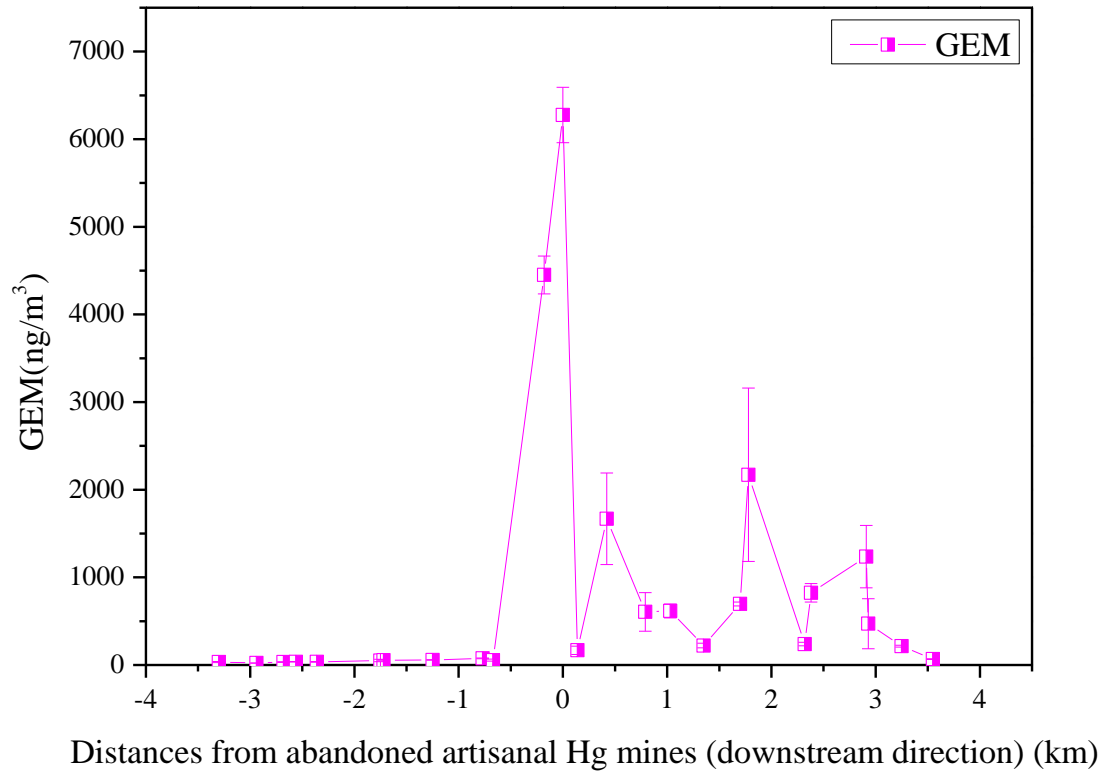


**Figure 3**

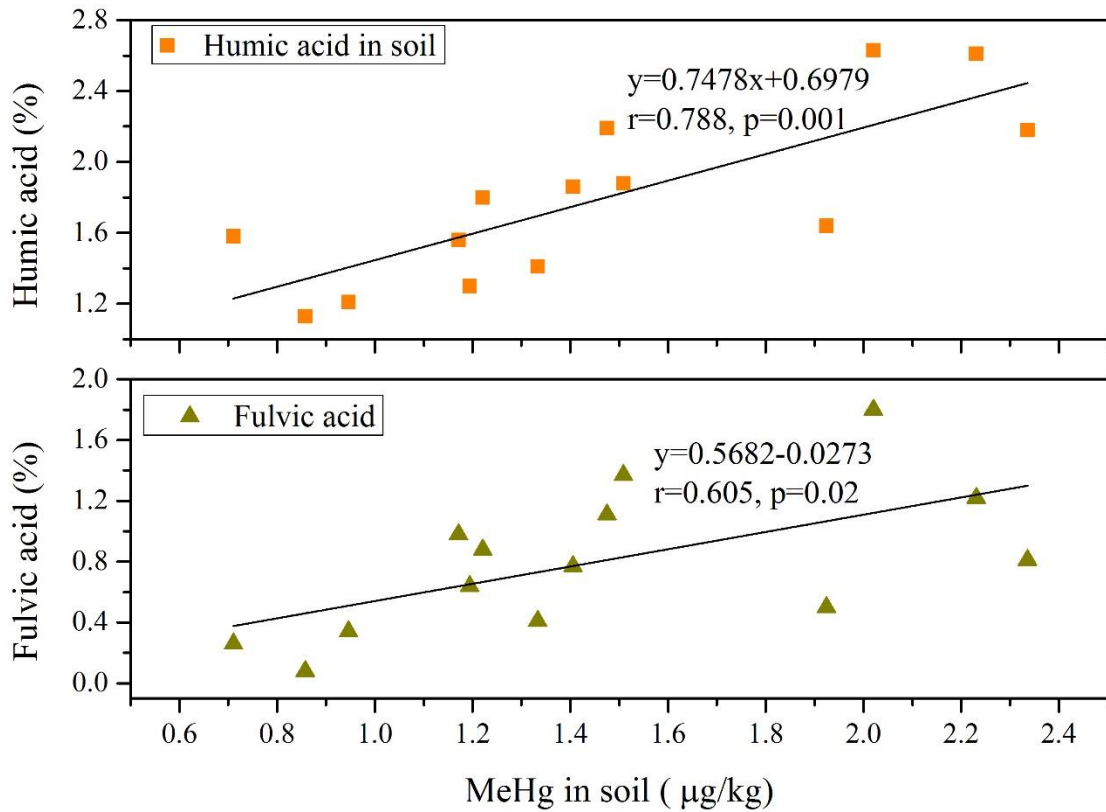


**Figure 4**





**Figure 5**



## Tables

**Table 1** Total mercury and methylmercury in surface waters from the YSK abandoned mining areas

**Table 2** The Hg and MeHg concentrations in surface water and soil collected from Hg mining regions worldwide

**Table 3** Total mercury and methylmercury in rice and soil gathered from the YSK abandoned mining areas

**Table 4** Correlations among soil THg (TH<sub>GS</sub>), soil MeHg (MeH<sub>GS</sub>), rice THg (TH<sub>GR</sub>), rice MeHg (MeH<sub>GR</sub>), and GEM in this study. Asterisk (\*\*) suggests correlation is significant at the 0.01 level, and (\*) denotes correlation is significant at the 0.05 level.

**Table 5** Correlations among soil MeHg, organic matter, humic acid, humilic acid, and fulvic acid in our research. Asterisk (\*\*) suggests correlation is significant at the 0.01 level, and (\*) denotes correlation is significant at the 0.05 level.

**Table 1**

Sample ID	Location	Unfiltered water		Filtered water	
		THg (ng/L)	TMeHg (ng/L)	DHg (ng/L)	DMeHg (ng/L)
<i>Upstream from Hg mines</i>					
Y01	Shileixi Creek	21	0.23	10	0.16
Y02	Shileixi Creek	28	0.17	15	0.17
Y03	Shileixi Creek	25	0.24	14	0.14
<i>Abandoned artisanal Hg mines</i>					

Y10	Shileixi Creek	24	0.55	10	0.35
Y11	Shileixi Creek	16	0.29	6.9	0.22
Y12	Shileixi Creek	13	0.33	4.7	0.17
<i>Downstream from abandoned industrialized Hg mines</i>					
Y13	Leachate from calcines	2390	0.43	473	0.30
Y14	Shileixi Creek	1004	0.53	61	0.31
Y15	Shileixi Creek	333	1.1	110	0.18
Y19	Shileixi Creek	214	0.39	19	0.31
Y20	Rongxi River	146	0.40	25	0.22
<i>Recommended Hg guidelines for water</i>					
Chronic effects to aquatic wildlife ( <a href="#">USEPA, 1992</a> )		770			
Headwater and natural reserves ( <a href="#">Chinese EPA, 2002</a> )		50			
Chinese drinking water Hg guideline ( <a href="#">Chinese MOH, 2006</a> )		1000			

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**Table 2**

Location	Unfiltered water		Filtered water		Soil		Reference
	THg	TMeHg	DHg	DMeHg	THg	MeHg	
	(ng/L)	(ng/L)	(ng/L)	(ng/L)	( $\mu\text{g/g}$ )	(ng/g)	
Hg mines, California, USA	0.65-45000	0.017-47	0.2-140	0.017-1.2	0.6-1500		Ganguli et al., 2000; Rytuba, 2000; Domagalski et al., 2001; Thomas et al., 2002; Nacht et al., 2004; Suchanek et al., 2009
Hg mines, Alaska, USA	0.1-2500	0.01-1.2	1.3-50		0.03-5326	0.02-41	Gray et al., 2000; Bailey et al., 2002
Almadén Hg mine, Spain	<D.L.-20300	0.04-30	2.5-136		3.4-8889		Higuera et al., 2003; Berzas Nevado et al., 2003, 2009; Gray et al., 2004; Millán et al., 2006; Bueno et al., 2009
Idrija Hg mine, Slovenia	0.20-708	0.01-0.6	0.03-134	0.10-0.27	0.18-2759	0.32-80	Hines et al., 2000; Gnamuš et al., 2000; Horvat et al., 2002; Gosar et al., 2006; Kocman et al., 2004, 2011
Palawan Hg mine, Philippines	1.0-31000	<0.02-3.1	8.0-30000		0.012-566		Gray et al., 2003; Maramba et al., 2006
Wanshan Hg mine, China	3.3-10580	0.012-25	1.1-655	<0.005-6.0	0.10-790	0.10-23	Horvat et al., 2003; Qiu et al., 2005, 2009; Zhang et al., 2010a, b; Søvik et al., 2011; Lin et al., 2010, 2011
Xunyang Hg mine, China	6.2-23500	0.022-3.7	0.21-3500	0.015-0.52	1.3-750	1.2-11	Zhang et al., 2009; Qiu et al., 2012a

Yanwuping Hg mine, China	3.8-51	0.20-2.7	0.85-27	0.038-0.38	0.24-240	0.34-7.3	<a href="#">Qiu et al., 2013</a>
Xiushan Hg mine, China	21-2390	0.17-1.1	4.7-473	0.14-0.35	0.48-68	0.21-3.7	<a href="#">This study</a>

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**Table 3**

Sample ID	Location	Soil		Rice	
		THg ( $\mu\text{g/g}$ )	MeHg ( $\text{ng/g}$ )	THg ( $\text{ng/g}$ )	MeHg ( $\text{ng/g}$ )
<i>Upstream from Hg mines</i>					
Y01	Shileixi Creek	0.48	0.88	4.8	3.0
Y02	Shileixi Creek	1.0	1.1	12	9.2
Y03	Shileixi Creek	4.9	1.3	14	7.8
Y04	Shileixi Creek	1.7	0.48	16	5.7
Y05	Shileixi Creek	3.0	0.25	10	5.3
Y06	Shileixi Creek	1.1	0.21	7.1	4.7
Y07	Shileixi Creek	1.7	0.29	14	5.8
Y08	Shileixi Creek	4.2	0.41	16	7.1
Y09	Shileixi Creek	5.6	1.1	18	12
<i>Abandoned artisanal Hg mines</i>					
Y10	Shileixi Creek	13	1.0	120	21
Y11	Shileixi Creek	68	3.7	384	64
<i>Downstream from Hg mines</i>					
Y17	Shileixi Creek	15	3.1	12	8.7
Y18	Shileixi Creek	2.6	2.2	34	9.3
Y19	Shileixi Creek	1.5	2.7	36	8.4
Y20	Rongxi River	2.4	1.6	14	9.2
<i>Recommended Hg guidelines</i>					
Probable effect concentration to dwelling organisms ( <a href="#">Macdonald et al., 2000</a> )		1.06			
Available for agricultural soil ( <a href="#">Chinese MOA, 2010</a> )		1.5			
Limit for cereal food ( <a href="#">Chinese, GB 2762-2012</a> )				20	

11 **Table 4**

Correlations					
	THgs	MeHgs	THgr	MeHgr	GEM
THgs	1				
MeHgs	0.655**	1			
THgr	0.965**	0.591*	1		
MeHgr	0.975**	0.621*	0.991*	1	
GEM	0.995**	0.728*	0.999*	0.993*	1

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15 **Table 5**

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Correlations					
	MeHgs	Organic Matter	Humic acid	Fulvic acid	Humin
MeHgs	1				
Organic Matter	0.388	1			
Humic acid	0.725**	0.342	1		
Fulvic acid	0.536*	0.119	0.838**	1	
Humin	0.385	0.4	0.362	-0.136	1

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