



Mercury pollution from artisanal mercury mining in Tongren, Guizhou, China

Ping Li^{a,b}, Xinbin Feng^{a,*}, Lihai Shang^a, Guangle Qiu^a, Bo Meng^{a,b}, Peng Liang^a, Hua Zhang^{a,b}

^aState Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China

^bGraduate University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Concentrations of total Hg (T-Hg) were measured in mine waste, stream water, soil and moss samples collected from the Tongren area, Guizhou, China to identify potential Hg contamination to local environments, which has resulted from artisanal Hg mining. Mine waste contained high T-Hg concentrations, ranging from 1.8 to 900 mg/kg. High concentrations of Hg were also found in the leachates of mine waste, confirming that mine waste contains significant water-soluble Hg compounds. Total Hg distribution patterns in soil profiles showed that top soil is contaminated with Hg, which has been derived from atmospheric deposition. Data suggest that organic matter plays an important role in the binding and transport of Hg in soil. Elevated T-Hg concentrations (5.9–44 mg/kg) in moss samples suggest that atmospheric deposition is the dominant source of Hg to local terrestrial ecosystems. Concentrations of T-Hg were highly elevated in stream water samples, varying from 92 to 2300 ng/L. Particulate Hg in water constituted a large proportion of the T-Hg and played a major role in Hg transport. Methyl-Hg (Me-Hg) concentrations in the water samples was as high as 7.9 ng/L. Data indicate that Hg contamination is dominantly from artisanal Hg mining in the study area, but the extent of Hg contamination is dependent on the mining history and the scale of artisanal Hg mining.

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1. Introduction

Mercury is one of the most severe environmental pollutants due to its high toxicity, easy uptake of its organic forms by biota, and the consequent accumulation in the food chain. Abandoned Hg mines worldwide are an environmental concern because Hg mining activities have resulted in significant Hg contamination to surrounding local environments. Furthermore, toxic Hg compounds present in discarded mine waste can easily be transferred to water, soil, and sediment, and thus, be converted to Me-Hg and accumulated in the food chain, posing a potential threat to local human health.

China contains abundant Hg resources and presently ranks as the third largest Hg producer in the world. The most important Hg production area is in Guizhou Province,

where cinnabar ore reserves represent approximately 78% of the total Hg in China (Qu, 2004). Large scale Hg mining activities in Guizhou ceased in the 2000s mainly due to economic reasons and environmental pollution concerns. However, demand for Hg has recently increased because Hg is utilized as a catalyst for polyvinyl chloride (PVC) plastic manufacturing from coal (NRDC, 2007). Demand for Hg has also increased as a result of restrictions of importation of Hg into China. Hence, Hg prices in China have increased sharply, which have stimulated the return of small scale and/or artisanal Hg mining in Guizhou Province.

In Guizhou, most of the artisanal Hg mining occurs in and around Hg mines, such as those in Laohugou, Banchang and Luoxi in the Wuchuan area, and Gouxu and Lao-wuchang in the Tongren area. Total Hg supply in China was estimated at approximately 1400 tons for 2004-nearly half of the global total. The Hg used in industry in China is supplied from four sources: legal mining, illegal mining, imports, and recycling. Since 2002, domestic Hg

* Corresponding author. Fax: +86 851 5891609.

E-mail address: fengxinbin@vip.skleg.cn (X. Feng).

production has increased steadily each year to meet China's internal demand, rising to an estimated 700 tons in 2004 (NRDC, 2007). The annual Hg production from the artisanal Hg mining in Gouxi and Laowuchang area was estimated to be 30 metric tons. Similar to the Wuchuan area (Li et al., 2008), most of the Hg ore (cinnabar) in Gouxi, Tongren was from numerous local, small scale Hg mines.

However, in Laowuchang Hg recovery is from abandoned mercuric chloride catalysts used in the manufacture of PVC plastic, where elemental Hg has been extracted from the catalysts. Generally, the catalyst is made of active C, containing about 8–12% mercuric chloride. If the mercuric chloride content is <5%, the catalysts are discarded. The abandoned catalysts are treated with lime, and then re-ported at 700–800 °C to recover elemental Hg. Unfortunately, during this process, a large portion of the elemental Hg is released to the ambient air due to poor Hg recovery processes and the lack of adequate environmental protection equipment (Li et al., 2006).

Studies conducted in Wuchuan showed that artisanal Hg mining has resulted in significant Hg pollution to surrounding environments (Qu, 2004; Qiu et al., 2006a) and Hg vapor exposure to the workers involved in mining (Iwata et al., 2007; Li et al., 2008). However, no studies of environmental Hg contamination and human Hg exposure have been carried out in Gouxi or Laowuchang, Tongren. In order to assess the environmental impact of artisanal Hg mining, T-Hg and Me-Hg concentrations were determined in mine waste, stream water, soil and moss samples collected from the Gouxi and Laowuchang areas. Additional studies are ongoing to evaluate exposure of Hg to workers involved in mining and local residents, using Hg analysis of hair, urine and food, but these results are not presented here.

2. Materials and methods

2.1. Study area

The Gouxi and Laowuchang study areas, are situated in the Tongren District, Guizhou Province, in SW China (Fig. 1). Tongren is a hilly and karstic terrain with elevations ranging from 205 to 1149 m. The region has a sub-tropical, humid climate characterized with abundant rainfall and mild temperatures. The annual mean precipitation is about 1200 mm and the annual mean temperature is 17 °C. The Gouxi and Longdong Rivers flow through the Gouxi and Laowuchang area, respectively, and then flow into the Jinjiang River and finally into the Yangtze River. The main soil types in both the Gouxi and Laowuchang areas are ultisol and limestone soil.

There are two small scale Hg mines located in the Gouxi River basin, but only one is in operation (Fig. 1). Similar to most of Hg mines worldwide, the primary ore in the mines is cinnabar; pyrite and other sulfide minerals, which are significant acid-water producing minerals, are rare. Gangue minerals are mostly dolomite, calcite and quartz. Most of the small Hg retorts in Gouxi are in the C1, C2 and C3 areas (Fig. 1), which are located along the banks of Gouxi

River because this was a favorable location for transport of Hg ore. Significant quantities of mine waste produced by these small Hg retorts were directly discarded into the stream channel.

Artisanal furnaces in Laowuchang were located in the C4 area (Fig. 1), which is upstream in the Longdong River basin. In this area, the Hg furnaces recovered elemental Hg from mercuric chloride catalysts, which were abandoned following the manufacture of PVC plastic. Waste products from artisanal mining were discarded into nearby drainages that discharge into the Longdong River.

2.2. Sample collection and preparation

Twenty-eight samples of mine waste and mine tailings were collected from discarded heaps in the C1, C2, C3 and C4 areas. Four soil profiles (SP1, 2, 3, and 4) at Gouxi and 3 soil profiles (SP6, 7 and 8) at Laowuchang were sampled (Fig. 1). Soil samples at different depths were collected using a self-regulating drill. One surface soil sample (S5) was collected in the C2 area. Naturally growing mosses that adhered to rock surfaces were also collected in the vicinity of the soil samples. Two moss samples from a control site were collected for comparison. At each sample site, the final moss sample was composed of 3–5 sub-samples. All samples were placed and sealed in clean polyethylene bags to avoid cross contamination.

Filtered and unfiltered water samples were also collected (Fig. 1). Water samples were collected from artisanal Hg mining sites or at the fluvial system confluence with its tributaries. Water samples were filtered in situ with a 0.45 µm polyvinylidene fluoride filter (Millipore). Prior to fieldwork, the borosilicate glass bottles were rigorously pre-cleaned by heating for 1 h in a muffle furnace at 500 °C. During collection, the filtration equipment and sample bottles were rinsed three times with the sampled water. Water samples were preserved by adding 0.4% (v/v) of distilled ultra-pure HCl within 24 h. Sample bottles were tightly capped and placed into double plastic bags to be stored in clean coolers (4 °C) until analysis. Collection, storage, and preservation techniques of samples strictly followed USEPA Method 1631 (USEPA, 1999).

In the laboratory, mine waste and soil samples were air dried, ground, and sieved to minus 100 mesh. The moss samples were washed with detergent and Milli-Q water in order to remove external Hg contamination and then air dried and ground. Water samples were preserved by refrigeration at 4 °C and were analyzed within 28 days.

Leaching tests of mine waste were performed by shaking 20 g of sample for 24 h end-to-end in 250 mL polyethylene centrifuge bottles with 200 mL of deionized water (pH 5.7). After centrifugation (2 h, 3000 rpm), all leachates were filtered through a 0.45 µm filter. Mercury species in solution were operationally defined as easily reducible (reactive) soluble Hg (ERS-Hg) and total soluble Hg (TS-Hg) (Biester et al., 1999).

2.3. Analytical methods

For T-Hg analysis, soil and mine waste samples were digested in a water bath using a fresh mixture of HCl and

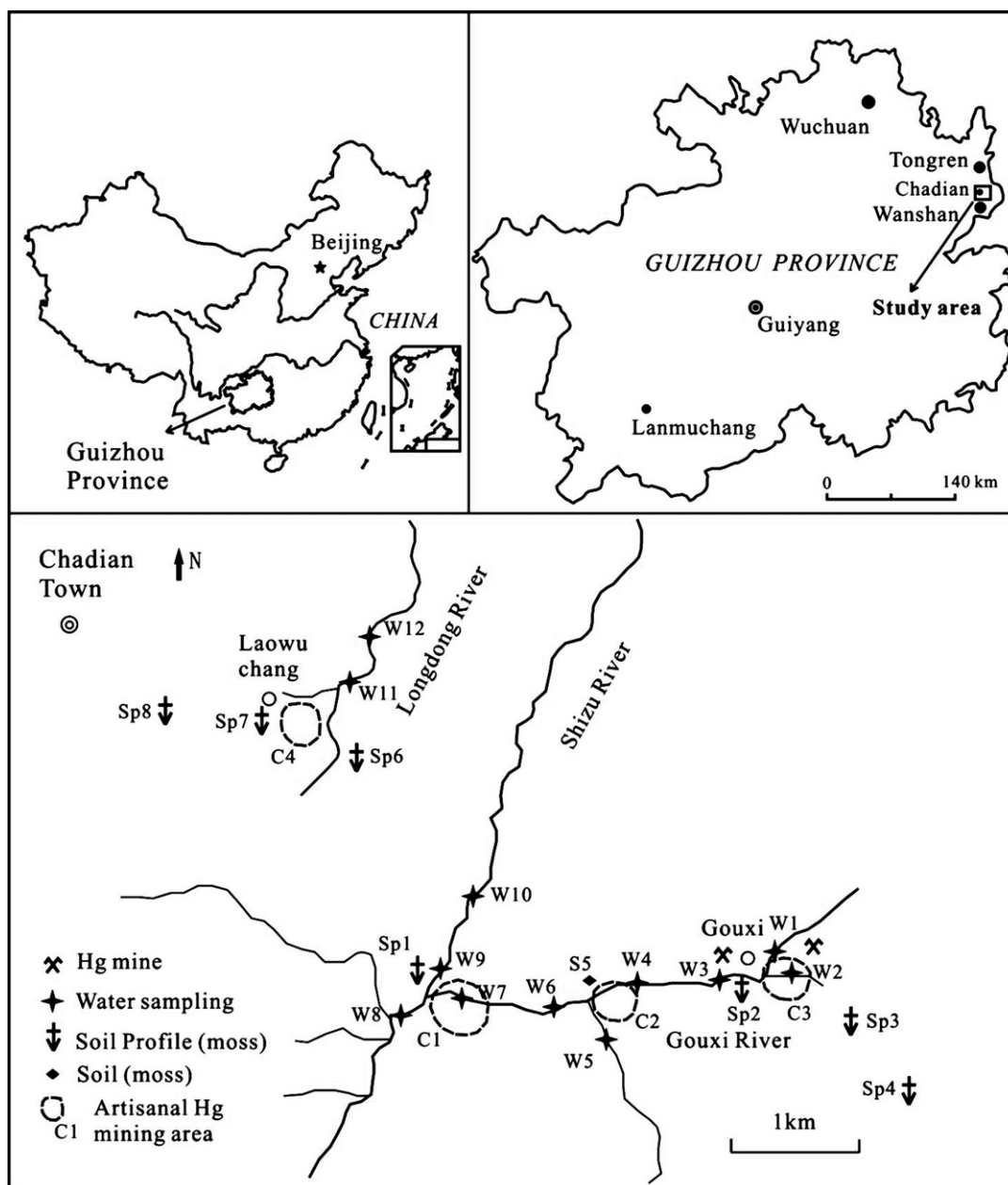


Fig. 1. Location of the sample sites in the Tongren area, Guizhou Province, China.

HNO_3 (1:3, v/v), whereas moss samples were digested with a mixture of HNO_3 and H_2SO_4 (4:1, v/v). Then T-Hg was measured on an aliquot of digested sample using cold vapor atomic absorption spectrometry (CVAAS).

Soil pH was measured using a pH electrode with a solid: water ratio of 1:2.5. Organic matter (OM) contents of soil samples were determined by the potassium dichromate volumetric method coupled with a water heating technique (Lu, 2000).

Operationally defined Hg species of reactive, dissolved and T-Hg in water samples were determined using a dual stage Au amalgamation method and CVAAS detection

(Yan et al., 2003; USEPA, 1999). Total and dissolved concentrations of Me-Hg in the stream water samples were determined using solvent extraction (Horvat et al., 1993), NaBH_4 ethylation, and GC-CVAFS detection according to USEPA Method 1630 (USEPA, 2001).

2.4. Quality control

Quality control consisted of method blanks, blank spikes, matrix spikes, certified reference materials and blind duplicates. The average T-Hg concentration of the geological standard GBW07405 was 0.30 ± 0.01 mg/kg

($n = 5$), which is comparable with the certified concentration of 0.29 ± 0.04 mg/kg. The percentage of recoveries on spiked samples ranged from 83% to 99% for Me–Hg in water samples. The relative percentage difference was <10% for T–Hg in soil, mine waste and moss duplicate samples as well as for T–Hg and Me–Hg in duplicate water samples. Limits of determination were 0.02 mg/kg for T–Hg in mine waste, soil and moss samples and 0.02 ng/L for T–Hg and 0.009 ng/L for Me–Hg in water samples.

3. Results and discussion

3.1. Mercury in mine waste

Concentrations of T–Hg in mine waste in the Gouxu area varied widely from 1.8 to 650 mg/kg (Table 1). Similarly, T–Hg concentrations in wastes from the abandoned catalysts in the Laowuchang area were also highly elevated (290 ± 390 mg/kg, ranging from 12 to 900 mg/kg, Table 1). Although the mine tailings samples from the C3 area exhibited the lowest T–Hg concentrations (23 ± 32 mg/kg), these were significantly higher than the baseline T–Hg concentration of 0.35 mg/kg in rocks reported in Wanshan area of China (Hua and Cui, 1994). Highly variable T–Hg concentrations in mine waste collected from the two areas probably indicates that retorting was not efficient, similar to results reported in other studies of Hg mines in the USA (Rytuba, 2003; Gray et al., 2004). Somewhat higher T–Hg concentrations in waste collected from the Laowuchang area was probably because the abandoned mercuric chloride catalyst contained about 2–5% of Hg (Xu, 1996), which was significantly higher than T–Hg in the Hg ores in the Tongren area (Qu, 2004).

During water leaching studies, highly variable TS–Hg concentrations were found in the leachates (Table 1). The mean of TS–Hg concentration in the leachates was $5.6 \mu\text{g/L}$ with a range of 0.004–130 $\mu\text{g/L}$. The most highly elevated TS–Hg concentrations ($27 \pm 56 \mu\text{g/L}$, ranging from 0.010 to 130 $\mu\text{g/L}$) were found in the leachates of residues from abandoned catalysts in the C4 area. This confirmed that the abandoned catalysts contained large quantities of soluble Hg. A significant correlation ($r = 0.73$, $p < 0.01$) between leachate Hg concentrations and Hg concentrations in the corresponding mine waste leached was found for samples collected from the two areas (Fig. 2). Hence the process of

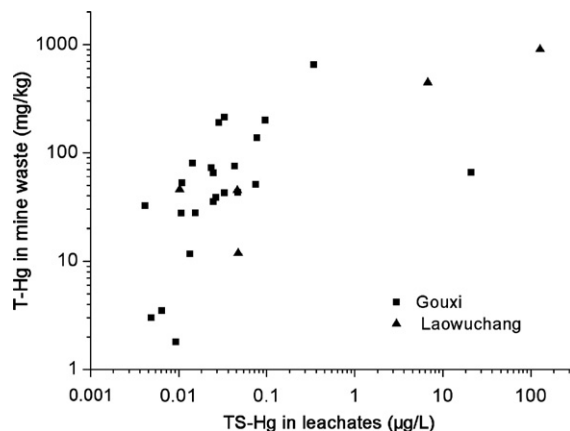


Fig. 2. The relationship between TS–Hg in leachates and T–Hg in corresponding mine waste in the Tongren area.

Hg retorting produced secondary by-product water soluble Hg compounds in the mine waste calcines (Kim et al., 2000; Biester et al., 1999).

Concentrations of ERS–Hg in the leachates varied from 0.003 to 95 $\mu\text{g/L}$ and were comparable to TS–Hg (Table 1). High concentrations of ERS–Hg could be bio-available for microbial transformation of inorganic Hg to Me–Hg, which could pose a threat to the downstream aquatic ecosystem because of the highly toxic nature of Me–Hg (Gray et al., 2004).

Comparisons of the T–Hg concentrations in mine waste from various studies worldwide are listed in Table 2. Concentrations of T–Hg in mine waste collected in the study area were comparable to the results of mine waste from the Wuchuan Hg mines, Guizhou, China (Qiu et al., 2006a; Li et al., 2006), Palawan Hg mine, Philippines (Gray et al., 2003), and Hg mines in SW Texas, USA (Gray et al., 2006). However, extremely high T–Hg concentrations, as high as 34,000 mg/kg, were reported in mine waste collected from the Almadén Hg mine, Spain (Gray et al., 2004). Highly elevated T–Hg concentrations in the mine waste and leachates in this study indicated that mine waste is a significant source of Hg contamination to the surrounding aquatic ecosystems in the Gouxu and Laowuchang areas.

Table 1
T–Hg in the mine waste and TS–Hg, ERS–Hg in the leachates

Area	Type	Hg concentration, mean \pm SD (range)		
		TS–Hg ($\mu\text{g/L}$)	ERS–Hg ($\mu\text{g/L}$)	T–Hg (mg/kg)
C1 ($n = 3$)	Mining residue	0.038 ± 0.033 (0.014–0.075)	0.016 ± 0.012 (0.006–0.029)	68 ± 15 (51–80)
C2 ($n = 6$)	Mining residue	3.5 ± 8.6 (0.011–21)	3.0 ± 7.2 (0.010–17.7)	87 ± 63 (28–189)
C3 ($n = 9$)	Mining residue	0.068 ± 0.11 (0.011–0.34)	0.028 ± 0.025 (0.006–0.086)	140 ± 205 (12–650)
C3 ($n = 5$)	Mining tailing	0.013 ± 0.017 (0.004–0.043)	0.008 ± 0.009 (0.003–0.024)	23 ± 32 (1.8–75)
C4 ($n = 5$)	Mining residue	27 ± 56 (0.010–130)	19 ± 43 (0.005–95)	290 ± 390 (12–900)

Table 2
Hg concentrations in mine waste and water from different studies worldwide

Location	Mine waste	Water		Authors
	T-Hg (mg/kg)	T-Hg (ng/L)	Me-Hg (ng/L)	
Wuchuan Hg mines, Guizhou, China	3.0–810	22–360	0.21–5.7	Qiu et al. (2006a) and Li et al. (2006)
Wanshan Hg mines, Guizhou, China	5.7–4400	17.3–10,580	0.012–0.766	Qiu et al. (2005), Horvat et al. (2003), and Feng et al. (2003)
Hg mines, Nevada, USA	1.9–2000	3.1–2000		Gray et al. (2002, 2003)
Au, Hg Amalgamation mining area, Nevada, USA		4.28–2107	0.305–7.2	Bonzongo et al. (1996)
Almadén Hg mine, Spain	160–34,000	7.6–13,000	0.41–30	Gray et al. (2004)
Idrija mine, Slovenia	42.6–1640	2.8–322	0.01–0.6	Hines et al. (2000) and Biester et al. (1999)
Palawan Hg mine, Philippines	28–660	8–31,000	<0.02–3.1	Gray et al. (2003)
Hg mines, California, USA	78–7240	2–450,000	0.003–47	Kim et al. (2004), Ganguli et al. (2000), Thomas et al. (2002), and Rytuba (2000)
Hg mines, SW Alaska, USA		1.0–2500	0.01–1.2	Gray et al. (2000)
Hg mines, SW Texas, USA	4.1–480			Gray et al. (2006)
Hg mines, Tongren, Guizhou, China	1.8–900	92–2300	2.6–7.9	This study

Table 3
Hg concentrations in stream water in the study area

Sample ID	T-Hg (ng/L)				Me-Hg (ng/L)			Total Me-Hg/T-Hg (%)
	Reactive	Dissolved	Particulate	Total	Dissolved	Particulate	Total	
1	90	35	320	360	2.5	0.10	2.6	0.72
2	38	96	1600	1700	3.4	4.5	7.9	0.47
3	23	110	1000	1100	4.2	2.8	7.0	0.64
4	7.5	84	230	310	4.3	1.7	6.0	1.9
5	5.1	20	72	92	3.0	1.5	4.6	0.90
6	18	88	2200	2300	3.3	3.3	6.5	0.29
7	5.4	23	740	760	4.2	0.058	4.3	0.56
8	9.1	110	400	510	3.4	3.1	6.5	17
9	9.0	23	330	360	3.6	1.1	4.7	1.3
10	23	24	230	250	4.8	2.1	6.9	2.8
11	170	250	2000	2200	4.9	1.1	6.0	0.27
12	22	210	1100	1300	2.9	1.7	4.6	0.36

3.2. Mercury in stream water

Concentrations of T-Hg were highly elevated in water samples and varied from 92 to 2300 ng/L (Table 3). The T-Hg concentrations in stream water samples collected in this study were highly elevated compared with the T-Hg (0.1–20 ng/L) of natural fluvial systems (Louis and Chamberland, 1995).

The particulate Hg concentration ranged from 72 to 2200 ng/L and constituted a large portion of the T-Hg concentrations in stream water collected in this study. This confirms that Hg tends to be bound to particulate matter in water. High concentrations of particulate Hg also indicates that most of the Hg transported downstream from the mines was as suspended material, probably particulate cinnabar remaining in mine waste. Particulate Hg concentrations sharply increased when water flowed through mine waste heaps (Fig. 3). Thus, mine waste is a significant source of Hg pollution to local environments in the Tongren area.

The reactive Hg and dissolved Hg concentrations in the stream water samples varied from 5.1 to 170 ng/L and from 20 to 250 ng/L, respectively (Table 3). Stream water collected from W11 (Fig. 1, Table 3) contains the highest reac-

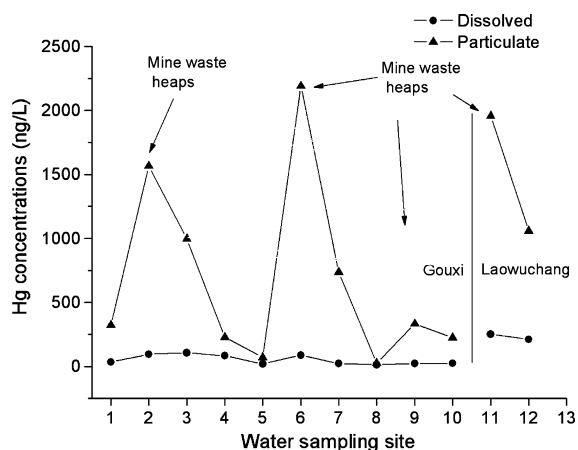


Fig. 3. Particulate and dissolved Hg concentrations in stream water collected in the Tongren area.

tive Hg and dissolved Hg concentrations and these results are probably due to leaching of highly soluble Hg in abandoned catalysts in the Laowuchang area where this sample was collected. The dissolved Hg concentrations determined

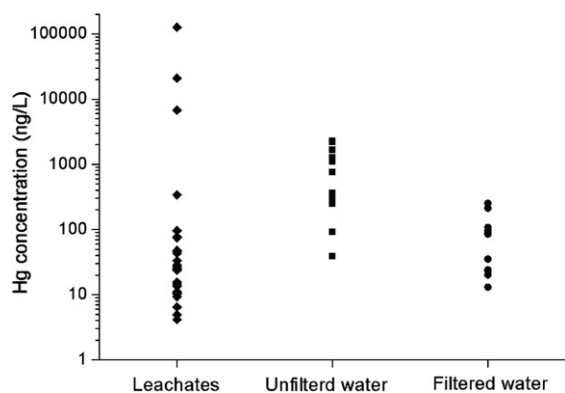


Fig. 4. Comparison of Hg concentrations from the leachates of mine waste and stream water samples in the study area.

in stream water in this study were comparable with the TS-Hg concentrations found for the leachates, except for the three highest concentrations (Fig. 4). High concentrations of dissolved Hg in stream water in Gouxu and Laowuchang suggest continuous release of soluble Hg from the mine waste.

Total Me-Hg and dissolved Me-Hg in water samples ranged from 2.6 to 7.9 ng/L and from 2.5 to 4.9 ng/L, respectively (Table 3). While particulate Hg is the dominant fraction of Hg in water, Me-Hg is the dominant dissolved fraction in the water samples. Dissolved Me-Hg accounted for 69.2% of total Me-Hg on average in the water samples, ranging from 43.4 to 98.6%.

The percentages of T-Hg as Me-Hg in stream water were generally <1%. However, higher percentages were found in the Gouxu River, ranging from 1.3% to 17% (sites 4, 8, 9, and 10; Table 3). No significant correlation between total Me-Hg and T-Hg concentrations were found in the water samples, indicating that environmental differences affect the transformation of Hg in the aquatic environments.

Concentrations of T-Hg in water from this study area were comparable to results (4.28–2107 ng/L) from elemental Hg lost to the Carson River during historic Au mining in Nevada, USA (Bonzongo et al., 1996), T-Hg concentrations (3.1–2000 ng/L) in the mine water runoff collected from Hg mines in west-central Nevada, USA (Gray et al., 2002; Gray, 2003) and T-Hg concentrations in stream water (1.0–2500 ng/L) downstream from Hg mines in SW Alaska, USA (Gray et al., 2000). An extremely high T-Hg concentration of 450,000 ng/L was found in a drainage downstream from a Hg mine in the California Coast Ranges, USA (Rytuba, 2000). Water flowing through Hg mine waste also contained high T-Hg concentrations (18,000–31,000 ng/L) at the Palawan Hg mine, Philippines (Gray et al., 2003). Compared to T-Hg concentrations in drainage from California and the Palawan Hg mines (Table 2), T-Hg results shown here are slightly lower, but the river in the study area has been contaminated by Hg from the surrounding artisanal Hg mining.

Me-Hg concentrations in stream water from in the Tongren area (2.6–7.9 ng/L, Table 2) were comparable to those from the Wuchuan Hg mines, Guizhou, China (0.21–5.7 ng/L,

Qiu et al., 2006a) and those due to elemental Hg contamination of the Carson River due to Au mining in Nevada, USA (0.305–7.2 ng/L, Bonzongo et al., 1996). However, extremely high total Me-Hg concentrations of 30 ng/L and 47 ng/L were reported in water samples at the Almadén Hg mine, Spain (Gray et al., 2004) and Hg mines in California, USA (Ganguli et al., 2000; Thomas et al., 2002), respectively.

3.3. Mercury in soil

Concentrations of T-Hg found in soil profiles generally exhibited patterns of increasing Hg near the surface (Fig. 5). Except for SP4, T-Hg concentrations in top soil (0–10 cm) were significantly elevated compared to soil >20 cm in depth. With increasing depth, T-Hg concentrations in soils decreased sharply and reached a stable concentration at the depth of 40–50 cm (e.g., 0.5 mg/kg, SP6, Fig. 5). A previous study has shown that artisanal Hg mining is a significant anthropogenic atmospheric Hg emission source (Li et al., 2006). Concentrations of T-Hg indicated that Hg in top soil was primarily from deposition of atmospheric Hg emitted during local artisanal Hg mining, and not from geological sources.

Concentrations of T-Hg in samples at the bottom of the soil profiles (0.2–1.0 mg/kg, Fig. 5) are interpreted to represent the local uncontaminated baseline concentration. This Tongren area T-Hg baseline concentration is comparable to that reported as the geochemical baseline in soil in the Wanshan area, China (0.1–1.2 mg/kg, Qiu et al., 2005). However, this T-Hg Tongren area baseline is slightly higher than T-Hg concentration reported for uncontaminated soil worldwide (0.01–0.5 mg/kg, Senesi et al., 1999). The high local baseline T-Hg concentration in the Tongren soil is related to the natural geologic sources of Hg in the study area, which is an area with numerous Hg deposits containing cinnabar.

Patterns of OM in most soil profiles show trends similar to those of T-Hg concentration (Fig. 5). High concentrations of OM were generally found in top soil, which decreased with depth. A significant correlation ($r = 0.64$, $p < 0.01$) between T-Hg and OM concentrations was observed in the soil profiles (Fig. 6). Organic matter has a strong capacity to complex and bind with Hg (Lindqvist et al., 1991; Biester et al., 2002). A positive correlation between organic C and T-Hg concentrations for soil samples has been previously reported (Hissler and Probst, 2006; Tomiyasu et al., 2003). Hissler and Probst (2006) concluded that most of the atmospheric Hg deposition was trapped by OM contained in soil. The amount and the quality of OM and the mechanisms regulating the partition of OM between aqueous and solid phases might play a major role in the distribution of T-Hg and in Hg transport through soil profiles (Semu et al., 1987). The accumulation of Hg in the organic-rich upper-soil layers and corresponding high T-Hg concentrations in the Tongren soil profiles is interpreted to be related to atmospheric Hg deposition.

High soil pH was found in the Tongren samples, varying from 6.2 to 7.5 with a mean of 7.0 (Figs. 5 and 6). No significant correlation between soil pH and T-Hg concentrations was observed (Fig. 6). Generally, high soil pH in the

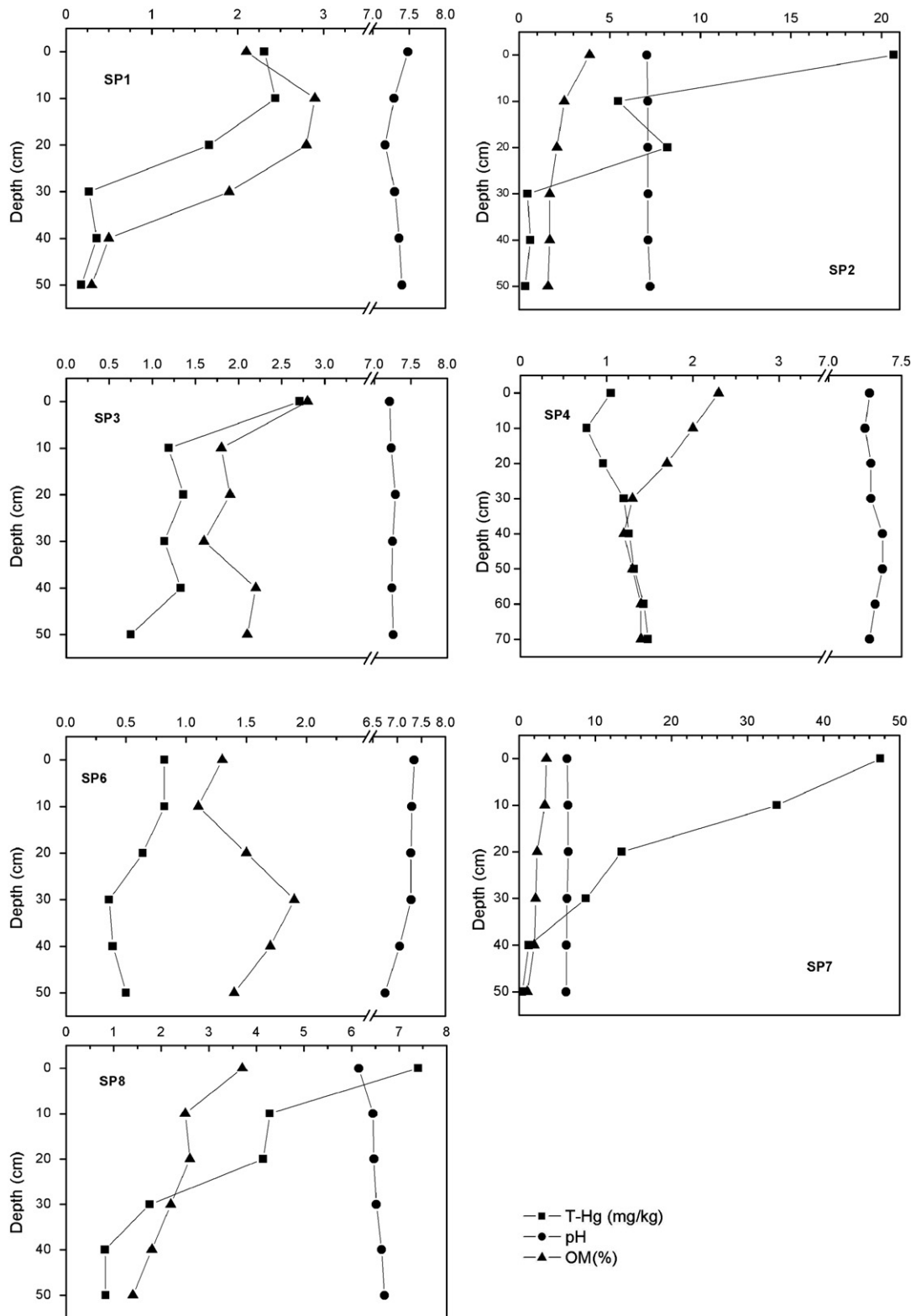


Fig. 5. T-Hg, OM and pH distribution patterns in soil profiles collected in the study area.

study area is related to soil mineralogy and geochemistry in the Tongren area. The bed rocks in the study area consist

dominantly of dolomite and limestone. The rocks contained a large quantity of alkali minerals and resulted in

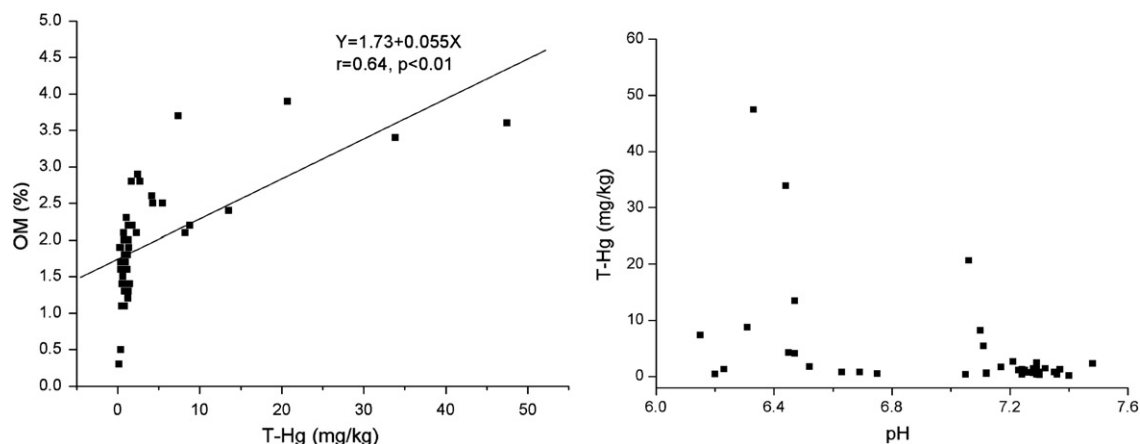


Fig. 6. The relationship between OM, pH and T-Hg concentrations in the soil samples collected from the Tongren area.

high soil pH. In addition, acid-water producing minerals, such as pyrite and other metal sulfides, are rare in the Tongren Hg ores which is another factor resulting in generally high soil pH.

Obviously, local soil Hg contamination has been influenced by the history and scale of Hg mining in the Tongren District. Concentrations of T-Hg in surface soil decrease significantly with increasing distance from the Hg mined areas. For instance at Gouxu, the highest T-Hg concentration in top soil was found in SP2 (21 mg/kg), which was the center of the artisanal Hg mining (Fig. 1). However, T-Hg concentration in the surface soil decreased to 2.7 mg/kg in SP3 and 1.1 mg/kg in SP4, which are sites located about 1 km and 2 km from SP2, respectively. Concentration of T-Hg was 3.1 mg/kg in soil sample S5 (Fig. 1, data not shown), which also indicates the effect of the artisanal Hg mining in the C2 area. In the Laowuchang area, the maximum T-Hg concentration in top soil was found in SP7, which is the center for Hg recovery from manufactured plastic catalysts. Spatial T-Hg distribution in the top soil could be attributed to atmospheric Hg sources related to emissions of Hg during elemental Hg recovery in the Laowuchang area.

Published T-Hg concentrations in soil from Hg mines in China are 5.1–790 mg/kg in Wanshan, 0.33–320 mg/kg in Wuchuan, and 8.4–850 mg/kg in Lanmuchang (Qiu et al., 2005, 2006a,b). At the Almadén mine, Spain, and the Idrija Hg mine, Slovenia, results for T-Hg in soil samples range widely from 6 to 9000 mg/kg and from 0.39 to 2759 mg/kg, respectively (Higuera et al., 2003; Gnamus et al., 2000). Concentrations of T-Hg in soil from the Gouxu and Laowuchang areas (0.18–47 mg/kg, Fig. 5) are much lower than those reported from these Hg mines. The lower T-Hg soil concentrations in the two areas may be due to the small scale and short term of Hg mining and retorting. However, compared with the Tongren baseline T-Hg concentration (0.20–1.0 mg/kg, Fig. 5), the soils in the study area were contaminated by the artisanal Hg mining.

3.4. Mercury in moss

Concentrations of T-Hg in the moss samples from the mining areas ranged from 5.9 to 44 mg/kg, which was sig-

Table 4

T-Hg concentrations in the moss samples collected in the study area and control site

ID	Species	T-Hg (mg/kg)
M1	<i>Anomodon minor</i>	25
M2	<i>Bryum tuberosum</i>	26
M3	<i>Dendrocypophorum decolyi</i>	44
M4	<i>Tortula schmidii</i>	22
M5	<i>Breidleria erectiuscula</i>	23
M6	<i>Rhynchostegium inclinatum</i>	17
M7	<i>Herzogiella renitens</i>	30
M8	<i>Brachythecium amnicolum</i>	5.9
M9 ^a	<i>Rhynchostegium inclinatum</i>	0.39
M10 ^a	<i>Breidleria erectiuscula</i>	0.63

^a Control site.

nificantly higher than those found in the control site (M9, 0.39 mg/kg; M10, 0.63 mg/kg, Table 4). The moss samples collected in the center of artisanal Hg mining (M2, M3, and M7) contained the highest T-Hg concentrations (26–44 mg/kg, Table 4).

Generally, naturally growing moss has been recognized as a useful bio-monitor of air pollution and has been widely used in tracing concentrations and deposition of airborne Hg (Bargagli et al., 1998; Steinnes et al., 2003). Qiu et al. (2005) found a strong positive correlation between T-Hg concentrations in moss and total gaseous Hg concentrations in the ambient air in Hg mined areas. Thus, elevated T-Hg concentrations in moss samples collected from the Tongren area indicated that the highly elevated total gaseous Hg in the ambient air and atmospheric Hg deposition is an important pathway of Hg contamination to local environments.

4. Conclusions

The results of this study confirmed significant Hg contamination of mine wastes, stream waters, soils, and mosses resulting from artisanal Hg mining in the Tongren District. Mine waste that contained high concentrations of T-Hg have resulted in Hg contamination to surrounding environments. Stream water was found to be highly con-

taminated with T-Hg and Me-Hg suggesting that Hg methylation was active in stream environments in the Tongren area. Top soils were also significantly contaminated with Hg, which was dominantly derived from atmospheric Hg deposition. Organic matter might play a major role in controlling the binding and transport of Hg in soil. Moreover, high T-Hg concentrations in moss collected in the Tongren area reflect the highly elevated total gaseous Hg concentrations in the ambient air, which is interpreted to be related to mining-related atmospheric Hg deposition in the study area.

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