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Mercury Mining in China and its Environmental and Health Impacts

Guangle Qiu, Ping Li, and Xinbin Feng

State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China

9.1 Introduction

Mercury (Hg) is a non-essential and toxic element. The primary metal Hg has been extracted from cinnabar ores. Because of the well established toxicity and human health effects of Hg as well as environmental concerns, the demand for Hg has decreased sharply since the end of the last century. In October of 2013, the Minamata Convention on Mercury was signed in Kumamoto, Japan; this treaty aims to protect human health and the environment from anthropogenic emissions and releases of Hg and Hg compounds.

China is rich in cinnabar ore resources and ranks the third in total Hg resources in the world. Large quantities of Hg mines are distributed throughout the Nation, especially in the southwest and central parts of China, including Guizhou, Shaanxi, Chongqing, Hunan, and Yunnan provinces. Among those provinces, Guizhou accounts for approximately 38.3% of the total cinnabar ore reserve in China. Owing to abundant cinnabar ore resources, China has a long history of Hg mining and Hg usage that may date back more than 2000 years. Most large-scale industrial Hg mining activities ceased in 2004; the Xunyang Hg mine, Shaanxi, is the only active one for metallic Hg production in China.

The presence of methylmercury (MeHg) in the environment is of great concern, because it is readily bioavailable and can be bioaccumulated and biomagnified in food chains. It is widely accepted that fish and its products are the dominant pathway of human exposure to MeHg. Recently, rice rather than fish has been verified as the major pathway of MeHg

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exposure to the residents living in Hg mining regions in China. The aim of this chapter is to review state-of-the-art knowledge on the distribution of Hg in the environment of Hg mining areas in China and its related environmental and human health impacts.

9.2 Mercury Mines and Mining

9.2.1 Mercury Mines

Three global mercuriferous belts have been discovered, circum-Pacific, Mediterranean-central Asia, and Mid-Atlantic Ridge. These belts are mainly distributed along global plate tectonic boundaries [1]. Most world large-scale Hg mines are distributed in those three belts, such as Almadén Hg mine, Spain; Idrija Hg mine, Slovenia; Monte Amiata Hg mine, Italy; New Almadén Hg mine, United States; Palawan Hg mine, Philippines; and Wanshan Hg mine, China.

China is located in the circum-Pacific global mercuriferous belt and has abundant cinnabar ore resources. Cinnabar ore reserves in China rank third in the world, after Spain and Italy. Approximately 31 Hg mines with cinnabar ore reserves more than 500 tons are found in China (http://www.chinamining.com.cn/report/default.asp?V_DOC_ID=1140). Most Hg mines are distributed in southwestern China, especially in Guizhou province, and the current total Hg reserves in Guizhou account for 38.3% of the total cinnabar ore reserve in China. The Wanshan Hg deposit, the largest in Asia and the third largest in the world in Hg reserves, lies in the eastern part of Guizhou province, which is considered to be the "Mercury Capital" in China.

9.2.2 Mercury Production

The primary metal Hg has mainly been extracted from cinnabar ores. Hylander and Meili [2] report that approximately a million tons of metallic Hg was produced during the past 500 years, more than 75 % from Europe and North America. The Almadén Hg mines in Spain alone contributed one third of the total metallic Hg production in the world.

Statistics showed that about 61,000 tons of metallic Hg were produced during the long history of Hg mining in China. The Hg mine in Wanshan, Guizhou, has alone contributed two thirds of the total reported national production. Different developing stages had different production of Hg in China. It developed rather rapidly in the Tang Dynasty (7th to 10th century), with the highest rate of development in history [3]. Large-scale Hg mining activities occurred in the 20th century. In the early 1960s, for instance, primary Hg production increased more than 2,000 tons. Since then to late 1980s production reached about 1,000 tons per year on an average. Annual Hg production in China decreased in the late 1990s to less than 500 tons, but has increased in recent years to more than 1,000 tons.

Even though Hg mining activities decreased and the regulations on the international trade of Hg became stricter in recent years, domestic Hg production has increased steadily each year since 2002 to meet the growing internal demand in China. According to the *Yearbook of Non-ferrous Metals Industry in China* 2010 (http://www.chinabookshop.net/yearbook-nonferrous-metals-industry-china-2010-p-10730.html), total mercury production in 2009 was estimated to be approximately 1,400 tons, more than half of the global total production.

9.2.3 Mercury Usage

In ancient China, cinnabar was used for red pigments, drugs, and preservatives. In old cities, such as Beijing, Nanjing, and Xi'an, red pigments can be observed in old constructions and former palaces. Ancient people also used metallic Hg by an amalgamation technique to extract gold.

Currently, the main areas of Hg demand in China are identified as acetylene polyvinyl chloride (PVC) production and the manufacture of thermometers and sphygmomanometers, batteries, and lighting. Jian et al. [4] reported that the total metallic Hg supply was about 1,563 tons in 2007. The PVC industry accounted for approximately of 60.1%, followed by manufacture of thermometers, 14.9%, batteries, 13.1%, sphygmomanometers, 6.27%, lighting, 4.93%, and others, 0.768% (Figure 1).

During the last 20 years, significant reduction in Hg use has occurred in the battery area. Total Hg demand was about 600 tons in 1995 and then decreased to less than 150 tons in 2008. However, the metallic Hg supply remained at about 1,000 tons per year, while global Hg production is about 2,000 tons annually [5]. China, therefore, plays an important role in controlling and reducing the global supply and trade of Hg.

9.3 Mercury in the Environment

Most large-scale industrial Hg mining activities recently ceased in China. However, abandoned Hg mines continue to impact the environment through the release of Hg-enriched mine-waste tailings (calcines), Hg-rich mine drainage, and Hg vapor. Therefore, highly elevated Hg concentrations were widely observed in air, soils, waters, and biota in Hg mining areas. Environmental concerns related to Hg mines are contamination of soils, sediments, and waters by mine wastes; Hg vapor released during ore processing and from mine tailings and contaminated soils; and Hg mine drainage release into watersheds. Generally, the large quantities of mine-waste tailings and heavily contaminated soils were considered the major sources for both soluble and particulate Hg in the surroundings.

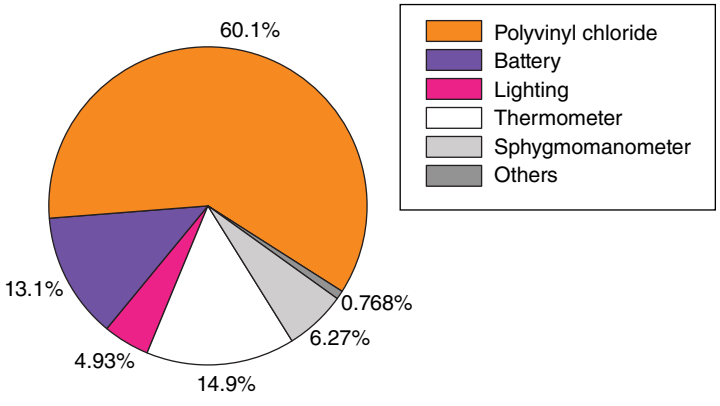

Figure 1 *Categories of Hg supply and usage in 2007 in China [4]. (See insert for color representation of the figure.)*

Most Hg mining is localized in remote regions where there is a lack of farmlands. The former farmlands occupied by Hg mining activities were reclaimed without any treatment after those Hg mines closed. The reclamation of contaminated farmlands increases the chance that Hg will enter into terrestrial food chains as well as the transformation of MeHg from inorganic forms under certain conditions. Hence, accumulation and biomagnifications of Hg, especially MeHg, in food chains poses a potential health risk to the residents in the region.

9.3.1 Air

9.3.1.1 Levels

The total gaseous mercury (TGM) concentrations in ambient air measured in Hg mining regions were characterized by significant variability and by extremely high values, ranging from 7.4 to 40,000 ng/m^3 in China (Table 1). The highest concentration of TGM, observed in Wuchuan Hg mining district, was related to artisanal Hg retorting activities [6]. The Wanshan Hg mining region also exhibited high TGM values of up to 2,100 ng/m^3 even after the Hg mines had ceased operation for more than five years.

TGM concentrations in ambient air in the Xunyang Hg mining region, the largest scale active Hg mine in China, were heavily impacted by anthropogenic activity. Elevated concentrations of TGM in ambient air were observed near the Hg retorting facility, reaching up to 410 ng/m^3 on average. Those high values decreased with distance from the retorting facility and reached a background value at 18 km in the downwind direction.

TGM concentrations in Hg mining regions were significantly elevated compared to the values reported for remote areas in China. Fu et al. [14–17] reported that TGM concentrations in ambient air at remote sites, such as Mt. Gongga, Mt. Changbai, Mt. Leigong, and Shangri-La, ranged from 1.6 to 4.0 ng/m^3.

Table 1 *Total gaseous mercury (TGM) concentrations in ambient air in Hg mining districts and remote regions in China*

Location	Time	Method	TGM (ng/m^3)	Reference
Wanshan, Guizhou	Nov 2002; Jul-Aug 2004	Tekran 2537A	18–1,100	[7]
	Feb 2003	Lumex RA-915+	30–1,950	
	Sep 2009	Lumex RA-915+	18–190	[8]
	May 2010	Lumex RA-915+	17–2,100	[9]
Yanwuping, Guizhou	Jul-Sep 2011	Lumex RA-915M	12–185	[10]
Wuchuan, Guizhou	Dec 2003; Dec 2004	Tekran 2537A	19.5–2,110	[11]
	Jun 2005	Lumex RA-915+	10–40,000	[6]
Lanmuchang, Guizhou	Dec 2002; May 2003	Tekran 2537A	24.5–210	[12]
Xunyang, Shaanxi	Sep 2009	Lumex RA-915+	7.4–410	[13]
Remote regions, China		Tekran 2538A	1.6–4.0	[14–17]

9.3.1.2 Emission Sources

Contributions from cinnabar ore retorting, widespread calcines, and heavily contaminated terrestrial and aquatic systems could represent major sources of Hg emissions to the atmosphere in Hg mining regions. Li et al. [18] found that the Hg emission factors during artisanal Hg retorting activities in Wuchuan ranged from 6.6% to 14.5% with an average of 9.8%, resulting in approximately 1.3–2.7 tons of Hg release per year into the atmosphere. These results showed that artisanal Hg retorting activity is an important atmospheric Hg source.

Wang et al. [7] conducted surveys on Hg emission flux from contaminated soils in Wanshan and found an average Hg emission flux of 162–27,827 ng/m^2/h and average Hg dry deposition flux of 0–9,434 ng/m^2/h, with average TGM concentrations of 17.8–1,100 ng/m^3 in ambient air. Even 10 years after the closure of Hg mining activities in the Lanmuchang Hg mining region in southwestern Guizhou province, Hg emission fluxes from contaminated soils reached up to 10,500 ng/m^2/h [12]. These results indicated that heavily contaminated soils are an important Hg emission source in abandoned Hg mining regions.

In addition, the mine-waste tailings generated by Hg mining and retorting activities have usually been emitted directly in the surroundings. Since the calcine is the ignited residue of cinnabar ores roasted at high temperatures, it contains a large quantity of secondary Hg minerals [19–23] that are readily released into the environment. In summer, calcines exhibited high emission of elemental Hg with a flux reaching up to 16,070 ng/m^2/h. This indicated that calcine piles are another important source of atmospheric Hg.

9.3.2 Mine-waste Tailings (Calcines)

The roasting process of cinnabar ores produced significant quantities of mine-waste tailings (calcines) that were directly piled nearby the adits or along the valleys and riverbanks. Calcines contain an abundance of soluble Hg compounds, such as elemental Hg, and Hg salts [24–26], resulting in release of large quantities of Hg into the surroundings via runoff, wind, and precipitation. Concentrations of total Hg in calcines differed widely in various Hg mining regions in China. Due to the inefficient and incomplete processing of cinnabar roasting, Hg concentrations in calcines ranged from 5.7 to 4,450 μg/g[27–29], comparable to the values reported by Gray et al. [20–23,30] and Bailey et al. [31].

These Hg-containing minerals generally have small particle sizes, i.e., micron or nanometer levels, and are easily attached to the surface of fine particles or dissolved in water in surface runoff. Thus, leachate of calcines generally contain elevated concentrations of Hg. For instance, the leachate of the calcines from Wanshan Hg mine can contain up to 4.46 μg/L of total Hg [32]. THg concentrations in calcines leachate of Almadén Hg mining district in Spain reached up to 120,000 μg/L, of which more than 70% was in dissolved Hg form [23].

Large amounts of soluble Hg compounds make calcines and its surroundings favorable sites for Hg methylation. Concentrations of MeHg measured in calcine samples collected from Wanshan Hg mining region ranged from 0.17 to 3.9 ng/g [27], comparable to the values found in Palawan Hg mining region but well below the 3,100 ng/g found in the Almadén Hg mining region, Spain [23][30]. Gray et al. [23] found that in calcines of the Almadenejos Hg mine, Spain the net Hg methylation rate is up to 11,100 ng/g/day, thousands of times higher than that of sediments, which were 1.7 ng/g/day. Generally,

environmental conditions such as high temperatures, wet conditions, abundant organic carbon, and high sulfur concentrations favor Hg methylation.

9.3.3 Soil

9.3.3.1 Levels

Surface soils affected by retorting activities in Hg mining regions in China exhibit a wide range from background levels to hundreds of μg/g in THg concentration in agricultural soils. Most samples at sites adjacent to calcines contained levels greater than 10 μg/g in soil in Yanwuping Hg mining regions [10], suggesting large impacts from those ignited residues. A significant negative correlation ($r = -0.73$, $p < 0.05$) of THg concentration and distance from the mine-waste tailings was observed in the Wanshan Hg mining region [33]. Bao et al. [34] reported that even 15 km away from the calcines tailings, THg in paddy soils still reached 15 μg/g, probably resulting from irrigation water that was heavily contaminated with Hg.

Soils in Hg mining areas were also found to contain elevated MeHg. The concentrations of MeHg in soil in the Wanshan mining areas ranged from 1.27 to 22.5 ng/g [35], and that in Wuchuan varied from 0.69 to 20 ng/g [28]. Similar to THg data, most elevated MeHg concentrations were observed at sites proximal to the calcine piles, indicating that calcine piles are potential sources of MeHg. Therefore, more attention should be paid to the calcine piles to reduce production of MeHg, and its drainages and leachates should be properly treated.

Generally, soil MeHg concentrations slightly increased with THg concentration increase ($r = 0.43$, $p < 0.0001$, Figure 2a), which indicates that the soil in Hg mining area favors Hg methylation and the concentration of bioavailable Hg species increases with the soil THg concentration. In the environment of Hg mining areas, the increase of bioavailable Hg concentration in soil might result from atmospheric Hg deposition. However, the contribution of atmospheric Hg in the soil to THg was much lower than that from calcines. A significant negative correlation was observed between THg concentration in soil and the percentage of MeHg in THg in soil ($r = -0.84$, $p < 0.0001$, Figure 2b).

The mercury methylation process in the environment depends on a number of factors, such as temperature, organic matter, soil pH, soil microbes, and redox conditions. Studies found that the change of seasons can also affect the net rate of Hg methylation: the peak rate of Hg methylation often appears in summer.

9.3.3.2 Spatial Distribution

The contour maps of Hg concentrations in soils collected from the Lanmuchang Hg mining district, Guizhou province [11,36,37] provide overall information on Hg dispersion, accumulation, and transport trends (Figure 3). Most of the highest THg concentrations were localized at numerous point sites, such as mining adits, retorting sites, and mine-waste tailings generated by historical artisanal Hg mining activities. Highly elevated concentrations of THg in soil observed near Hg mine-waste tailings and Hg concentrations in soil rapidly decreased beyond 4–8 km from the tailings. The southeast direction of the Qingshui River upstream, which flows through the mining district, seems to be the dominant trend in Hg dispersion.

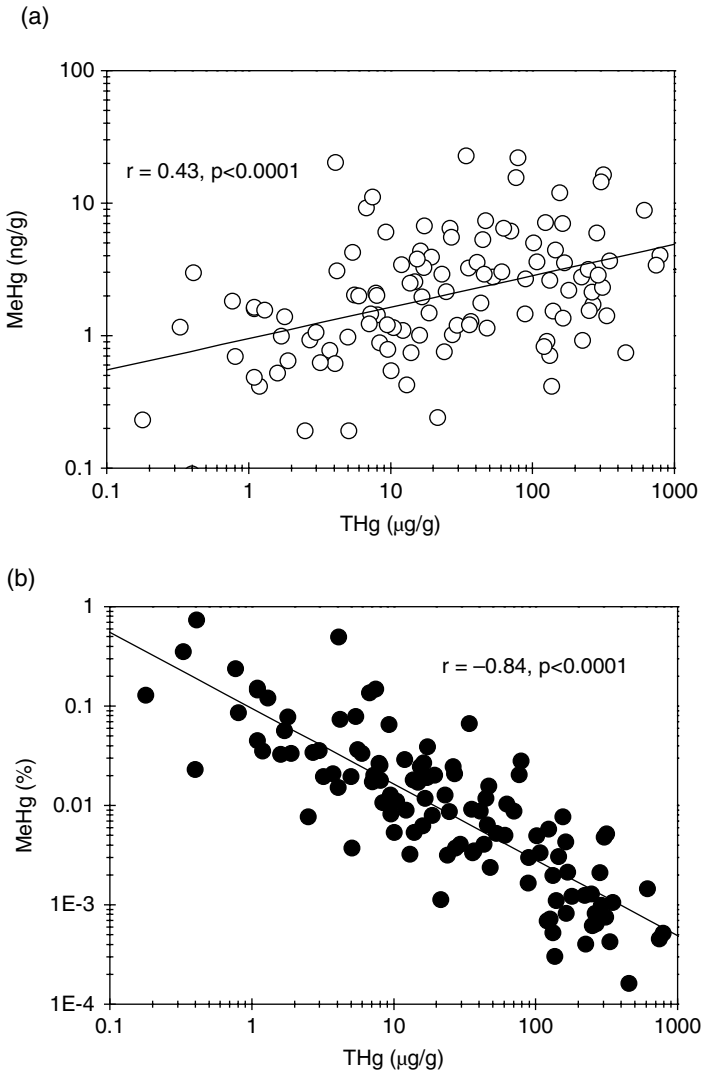

Figure 2 *Relations among THg, MeHg and its ratios in Hg mining areas in China*

In contrast to THg distribution in soil, the highest MeHg concentrations in soil were scattered along creeks and could be found upstream, midstream, and downstream of the tailings, indicating that methylation occurs readily in the soil along the river banks. Flooded paddy soil is an active net Hg methylation site. Typically, the soil near the streams on both banks floods readily and the anaerobic environment generated in the paddy soil is favorable to Hg methylation. At the same time, the moist environment is very conducive to microbial activity, resulting in high MeHg levels in soil along the banks of the river.

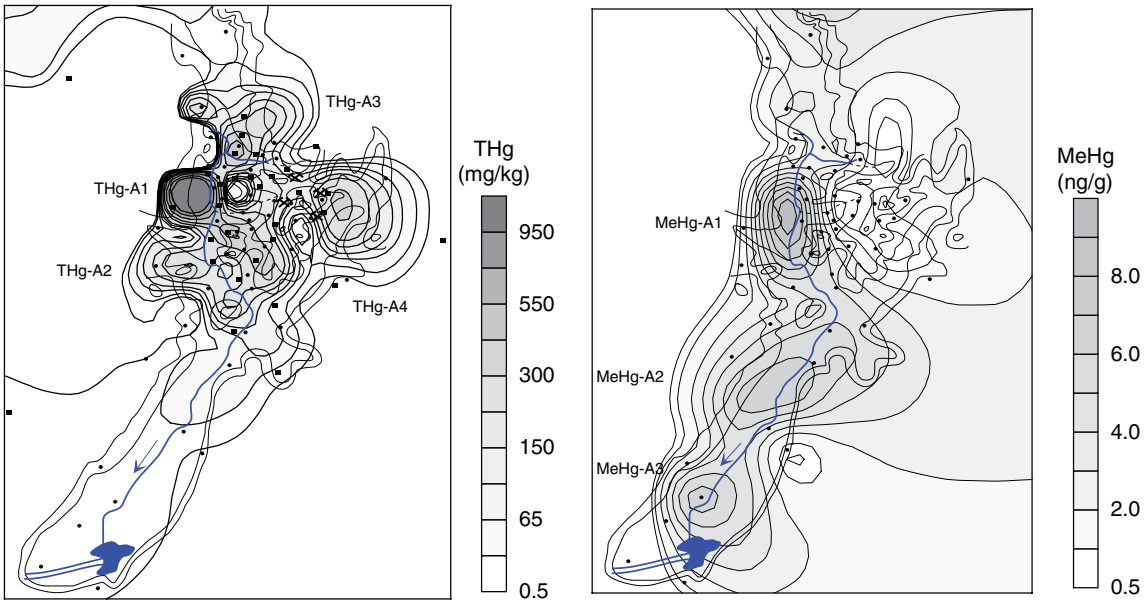

Figure 3 *Spatial distribution of THg (left) and MeHg (right) in surface soil with sampling locations in the Lanmuchang Hg mining district, China [37]. Reprinted from [37] with permission of Elsevier. (See insert for color representation of the figure.)*

9.3.3.3 Bioavailability

THg concentrations in soil reflect the degree of soil pollution, but do not indicate the potential environmental risk of Hg pollution [38]. Speciation information for soil Hg can indicate how its bio-toxic effects depend on its speciation in environmental media [39]. Using a modified sequence extraction procedure, we investigated the speciation of Hg in soil and sediment in Wanshan Hg mining areas. The total leachable Hg concentrations in sediment and soil reached up to 480 μg/g and 130 μg/g, respectively. The exchangeable Hg and strongly-bound Hg accounted for 10–30% and 20–40% of the total Hg [34,40]. Although soluble Hg and exchangeable Hg accounted for a small portion of THg, the absolute concentrations of these species in polluted soil were much higher than those found in soil at reference sites.

9.3.4 Water

Surface water affected by Hg mining contained elevated THg concentrations, ranging between 6.2 and 23,500 ng/L in China (Table 2). Peak concentrations of THg in surface water were generally found at sites directly affected by leachates from the calcines piles, verifying that calcine piles are important sources of Hg in surface waters. Horvat et al. [35] found that the THg concentration in leachate in Wanshan calcines was up to 10,580 ng/L, of which dissolved Hg (DHg) concentrations were in the range of 300–1,900 ng/L. In Xunyang Hg mining region, the leachate displayed extremely high THg levels of 23,500 ng/L [41]. Generally, however, the high THg concentrations show the characteristics of high particulate Hg levels, suggesting that most of the Hg transported downstream from the Hg mines occurred in suspended matter, probably as cinnabar particles.

Since the building of the Wukeng settling pond in the Wanshan Hg mining region, total Hg concentrations in the pond water were continuously monitored from 2000 to 2012; the resulting data are shown in Figure 4. Generally, THg concentrations showed a decreasing trend with values varying between >5.0 μg/L and 0.5 μg/L. In 2000, THg concentrations in

Table 2 *Mercury concentrations in surface water in Hg mining areas worldwide (ng/L)*

Location	THg*	DHg**	TMeHg***	References
Wanshan Hg mines, China	15–10,600	11–1,900	0.31–25	[32,35]
Tongren Hg mines, China	92–2,300		2.6–7.9	[42]
Wuchuan Hg mines, China	22–362	8.2–75.7	0.21–5.7	[28]
Lanmuchang Hg mines, China	25–7,020	<3–1,300		[37,43]
Xunyang Hg mine, China	6.2–23,500	0.21–3,500	0.022–3.7	[13,41]
Almadén Hg mines, Spain	7.6–20,300		0.41–30	[23,44]
Idrija Hg mines, Slovenia	2.8–322	0.03–134	0.01–0.6	[45,46]
Alaska Hg mines, USA	0.1–2,500	1.3–50	0.01–1.2	[20]
California Hg mines, USA	2–45,000	0.2–140	<0.003–47	[19,47–49]
Nevada Hg mines, USA	2.1–2,107	2–56	0.039–0.92	[50]
Palawan Quicksilver Mine, Philippines	120–31,000	8–30,000	<0.02–3.1	[30]

* Total mercury concentration
** Dissolved mercury concentration
*** Total methylmecury concentration

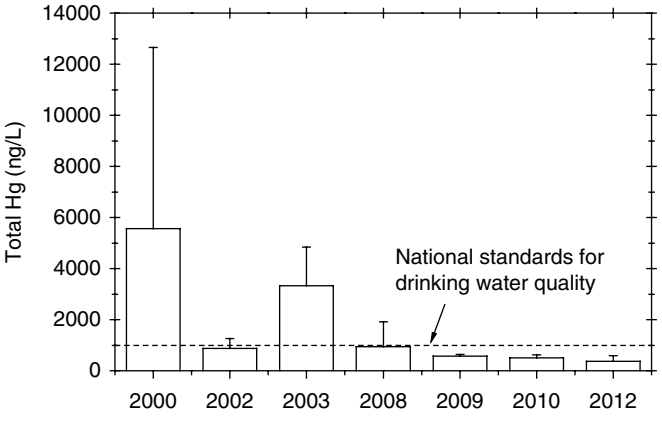

Figure 4 *Concentrations of THg in the water collected from Wanshan Hg mine Wukeng settling pond for the last 12 years [32,35,52]*

water exceeded 5.0 μg/L. The values decreased to <1.0 μg/L in 2002, but increased to 3.0 μg/L as a result of a rainstorm in 2003. In 2010, the values reached 0.5 μg/L, below the guideline value for National Standards for Drinking Water Quality set by the Ministry of Health, People's Republic of China [51].

Surface water in Hg mining regions also contained high concentrations of MeHg, reaching 25 ng/L [32]. The MeHg concentration in water showed a positive correlation with the dissolved Hg concentration in water, suggesting that MeHg in surface water mainly existed and migrated in the dissolved form, which is different from THg [53]. Affected by calcines leachates, the surface water environment favoured Hg methylation processes. The high MeHg in water adjacent to calcines would likely become a new pollution source for the downstream ecosystems.

9.3.5 Biota

9.3.5.1 Fish

Since fish are rare in streams of most Hg mining regions in China, few data on Hg concentrations in fish are available. Qiu et al. [32] reported that THg and MeHg concentrations in fish muscles collected from Wanshan Hg mining region were in the range of 0.061–0.68 μg/g and 0.024–0.098 μg/g, respectively. Similar results were observed in the Wuchuan Hg mining region with a range of 0.019 to 0.24 μg/g for THg and of 0.0061 to 0.11 μg/g for MeHg. The average THg concentrations in fish were 0.29 μg/g in Wanshan and 0.086 μg/g in Wuchuan, respectively, both of which values did not exceed the nation's guideline for acceptability of 0.50 μg/g.

Results for THg in fish from Wanshan were comparable to those of fish collected from Alaska Hg mines, but MeHg concentrations were much lower [23]. High average ratios of MeHg to THg (expressed as percent) were generally more than 90% in Alaska, while these ratios were as low as 28% and 41% on average in Wanshan and Wuchuan, respectively. Similar low MeHg-to-THg ratios were also reported for fish inhabiting areas impacted by Hg-contaminated sediments [54].

Obviously, factors such as species, age, sex, and feeding habits might impact MeHg to THg ratios. A weakly positive correlation ($r = 0.37$, $p < 0.5$) observed between MeHg and THg in fish from Wanshan and Wuchuan Hg mining areas might suggest that factors besides Hg concentration are influencing MeHg concentrations in these fish. Given the limitations of available data, however, reasons for the unusually low MeHg fraction in fish from Hg mining regions in China remain uncertain.

9.3.5.2 Rice

Rice is a staple food in China as well as in other regions of Asia. Rice samples collected from Hg mining regions in China exhibited a wide range of 9.0–1,120 ng/g for THg. Compared to the National Permitted Limit of 20 ng/g for THg in cereals in China, THg concentrations in rice samples collected from Hg mining regions were highly elevated. This demonstrated that rices cultivated in Hg mining areas were heavily contaminated with Hg because of the historic mining and retorting activities there.

Rice was also reported to accumulate higher levels of MeHg than other grains [55]. Compared to the edible parts of other crops, MeHg concentrations are observed to be much higher in rice, showing rice's high ability to accumulate MeHg.In the rice paddies in Hg mining areas, for instance, MeHg concentrations in grain can reach up to 170 ng/g, 1 to 2 orders of magnitude higher than MeHg in paddy soil. In rice, MeHg can account for more than 90% of THg, but the highest MeHg concentration in corn in the same area is only 1.3 ng/g.

MeHg bioaccumulation factors (BCFs) in over 97% of the rice samples tested were higher than 1 with an average value of 5.5–5.6, while inorganic Hg BCFs of rice, similar to other plant species, were generally much lower than 0.5. Data show that rice MeHg BCF values were 3 to 4 orders of magnitude higher than inorganic Hg [52]. Since MeHg is more easily translocated than inorganic Hg forms into above-ground plants, elevated MeHg concentrations in paddy soils are the major source of MeHg in rice [56]. Due to its high ability to accumulate in rice, MeHg in the terrestrial food chains has received considerable attention [52,57].

The phenomenal ability of MeHg to be accumulated in agricultural plants is rarely reported in the literature. Studies have shown that at the soil-plant root interface a layer of "iron plaque" exists that can effectively prevent soil inorganic Hg and MeHg ions from entering into the plant. In Hg mining areas, high MeHg concentrations can be detected in rice seeds and MeHg BCF values for rice seeds were usually higher than 1. These results showed that rice is a hyper-accumulator for MeHg.

9.3.5.3 Other Crops

In Hg mining areas, other crops such as corn, tobacco, and vegetables also suffered significantly from Hg pollution. THg concentrations in cabbages grown in the Lanmuchang Hg mining area, Guizhou, can reach as high as 18,000 ng/g, significantly above the Chinese National Food Standard of 10 ng/g for total Hg. Uptake of Hg in plants was primarily through leaves rather than through the root system. In Hg mining areas, TGM concentrations in ambient air are elevated due to Hg emissions from calcine piles and Hg-contaminated soils. THg concentrations in the leaf tissues of plants in the downwind direction from contaminated sites were much elevated.

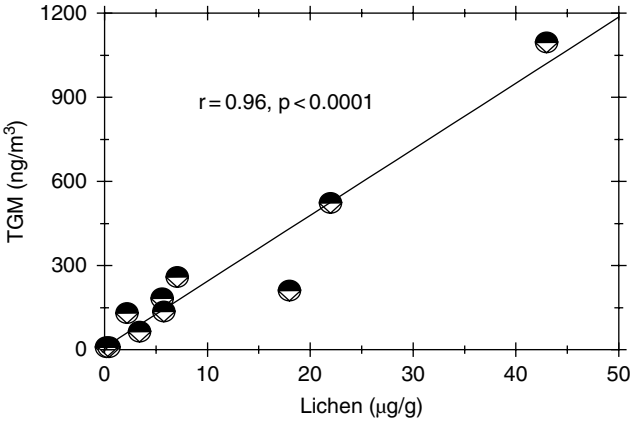

Figure 5 *Correlation between THg concentration in lichen and total gaseous Hg concentrations in ambient air. Reprinted from [37] with permission of Elsevier*

Lichen and moss are considered to be good biomarkers of air pollution. The Hg concentration of natural lichen grown on rock surfaces in the Wanshan Hg mining region reached up to 95,000 ng/g. Generally THg concentrations in lichens were significantly elevated at sites close to the Hg smelters and calcine tailings compared to sites far away from the contamination sources. Because nutrients of lichen are mainly from dry and wet deposition, the main source of Hg in lichen is also from atmospheric Hg deposition. THg concentrations in lichen were positively correlated with TGM concentrations in ambient air (Figure 5). The results also demonstrated that natural lichen plants on rock surfaces can serve as good atmospheric Hg pollution indicators.

9.4 Human Exposure and Health Risk Assessment

9.4.1 Human Exposure

As shown in Figure 6, in Wanshan Hg mine Li et al. [58] demonstrated a gradient of blood THg and MeHg concentrations at different sites with the distance from the pollution source in the two catchments. The highly elevated THg and MeHg levels in blood samples were observed in the upstream region (sites A, D, and E), which were seriously impacted by mine wastes. Blood THg and MeHg levels at site C and G basically represented the regional background level. In the total population, the overall mean of blood THg concentration was 12.2±15.0 μg/L (2.15–30.8, 95% confidence interval). The average of blood MeHg at different sites ranged from 2.20 to 9.36 μg/L. MeHg constituted 52.8±17.5% of THg in all blood samples on an average. About 72.6% (122/168) of blood THg concentrations exceeded 5.8 μg/L, which is the blood Hg level equivalent to the current RfD set by USEPA.

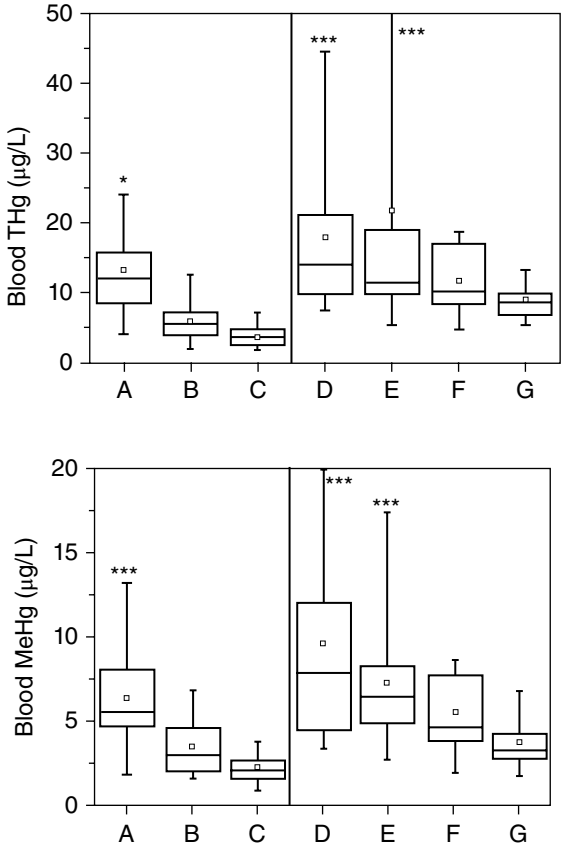

Figure 6 *Comparison of blood Hg in the population from Wanshan Hg mine. Adapted from Li et al. (2015) with permission from American Chemical Society*

9.4.1.1 Hair

Hair concentrations of THg and MeHg and percentages of THg as MeHg in the population from different Hg mines are summarized in Table 3. The average of hair THg for the control group was 0.78±0.28 μg/g [59], which is in the range considered "normal" by WHO (2 μg/g). Highly elevated THg concentrations were found in workers involved in artisanal mercury mining (AMM), with averages of 25.7±24.0 and 47.7±75.1 μg/g reported for workers in Wuchuan and Tongren Hg mines, respectively. Local residents in the Hg mine area also showed elevated hair THg concentrations, with average values of 5.04±7.57, 3.04±3.00, and 7.89±2.85 μg/g for residents in the Wanshan, Wuchuan, and Tongren Hg mines, respectively.

The average hair MeHg value for the control group was 0.65±0.25 μg/g [59]. In Tongren Hg mining areas, the average hair MeHg concentrations were 5.24±2.80 and 3.76±1.73 μg/g for AMM workers and local residents, respectively. Previous studies confirmed that rice can accumulate high levels of MeHg (170 ng/g) in Hg mining areas [55] and that rice, rather than fish, is the main route of human MeHg exposure in the Wanshan Hg mining area

Table 3 *Comparison of human hair Hg concentrations in different Hg mines*

Site	Subject	*n*	THg* (μg/g)	MeHg** (μg/g)	MeHg/THg (%)	Reference
Wanshan	Residents	95	5.04±7.57 (0.60–58.5)	1.85±1.16 (0.23–5.55)	55.3±22.5 (4.27–98.7)	[57]
Wuchuan	Workers	38	25.7±24.0 (2.03–93.1)	1.22±0.54 (0.52–2.53)	13.5±20.1 (1.10–96.8)	[60]
	Residents	100	3.04±3.00 (0.49–23.1)	1.25±0.74 (0.45–4.21)	53.7±27.8 (5.80–99.9)	
Tongren	Workers	18	47.7±75.1 (4.80–326)	5.24±2.80 (2.35–10.6)	28.5±23.3 (2.93–74.1)	[61]
	Residents	25	7.89±2.85 (3.52–13.1)	3.76±1.73 (1.87–9.23)	50.2±18.6 (23.6–99.3)	
Changshun	Residents	40	0.78±0.28 (0.32–1.72)	0.65±0.25 (0.26–1.38)	83.5±12.6 (52.7–99.9)	[59]

* Total mercury concentration
** Methylmercury concentration

[57], Guizhou Province [52] and the inland area of southern China [6]. The highly elevated hair MeHg levels indicated significant potential human health risk in Hg mining areas.

Considering the percentages of THg as MeHg in hair samples, the AMM workers showed the lowest ratios, with means of 13.5±20.1% and 28.5±23.3% for workers in Wuchuan and Tongren, respectively. Hair THg concentrations are considered a good biomarker of human MeHg exposure. For fish-eating populations, MeHg is the main form in the hair and can constitute from 80% to 98% of THg. Therefore, THg concentrations in hair samples are widely used as proxies for MeHg concentrations in human MeHg exposure. In this study, hair MeHg constituted only about 20% of THg on average for AMM workers, while the percentages of THg as MeHg in hair samples were about 50% for the local residents. As well, the ratios varied significantly for individuals.

Significant correlations were found between hair THg and UHg for AMM workers [59] and between inorganic mercury (IHg, which is defined as total Hg substrate MeHg) in hair and total gaseous mercury (TGM) concentrations in the ambient air [60] in Wuchuan Hg mine. The results from a simulated experiment indicated that the increases of hair THg concentrations were less than 1 μg/g under direct exposure of Hg vapor, which was negligible compared with hair THg concentrations in occupationally exposed workers [62]. The inhalation of Hg vapor constituted the major fraction of IHg exposure for the AMM workers [61], which resulted in highly elevated hair IHg concentrations. For the local residents, consumption of vegetables, rice, and meat accounted for >90% of the IHg exposure [63].

9.4.1.2 Blood

In the Chatian Hg mine, the mean Hg concentration in blood of the local residents was 6.09±3.26 μg/L (Table 4), significantly above those in the control area (3.67±0.82 μg/L [64]). The exposed groups in Wanshan Hg mine were characterized by high serum Hg concentrations (mean of 38.5 μg/L), again highly elevated compared with that of the control groups (mean of 0.91 μg/L) [65].

Table 4 *Comparison of blood Hg (B-Hg) concentrations in the population from Hg mines*

Site	Subject	*n*	B-Hg		Reference
			Mean±SD	Range	
Chatian	Local residents	54	6.09±3.26	1.29–15.07	[64]
	The control group	47	3.67±0.82	0.97–5.06	
Wanshan [a]	The Exposed group	37	38.5±61.5	1.85–210.3	[65]
	The control group	35	0.91±0.28	0.41–1.20	

[a] Serum Hg.

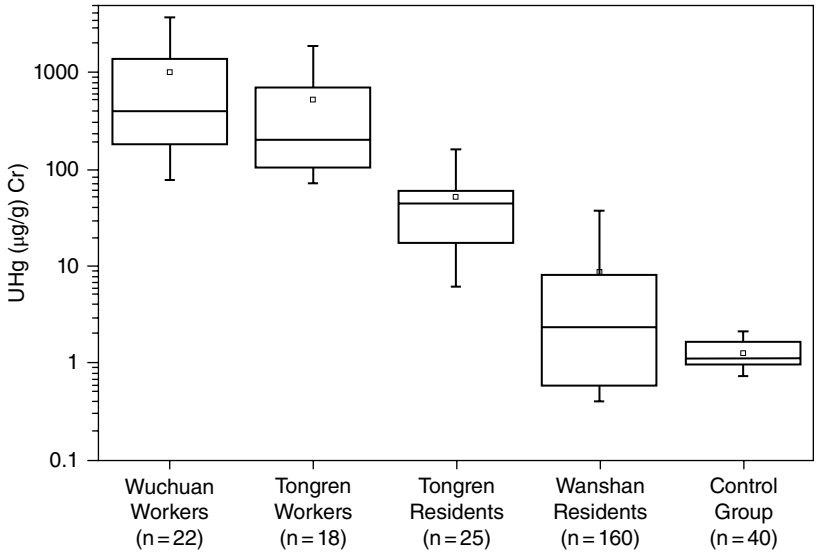

Figure 7 *Comparison of UHg in the population from different Hg mines in China. Data from [59,61,63] with permission of Elsevier*

9.4.1.3 Urine

Comparisons of total Hg concentrations in urine (UHg) in different populations from Hg mines are shown in Figure 7. The geometric mean of UHg for the control group was 1.30 μg/g creatinine (Cr) with a range of 0.68–2.32 μg/g Cr. However, the geometric mean for workers involved in artisanal mercury mining (AMM) in the Wuchuan Hg mine was 463 μg/g Cr, which was elevated by two orders of magnitude compared to that of the control group. The geometric means of UHg were 297 and 35.1 μg/g Cr for the AMM workers and the local residents from Tongren Hg mine, respectively. The geometric mean of UHg was 3.04 μg/g Cr for the local residents in Wanshan Hg mines. The maximum UHg concentration for occupational workers recommended by WHO [66] is 50 μg/g Cr and UHg levels for the general population should not exceed 5 μg/g Cr [67]. The highest UHg levels in workers involved in AMM from Wuchuan Hg mine reached 6,150 μg/g Cr, which is about 120 times higher than the occupational exposure limit level [66]. In conclusion,

workers involved in AMM in Guizhou Hg mines are seriously exposed to IHg and the local residents are also exposed to IHg at a significant level.

Previous studies confirmed that AMM can emit a significant amount of Hg vapor to the ambient air [18], resulting in significant Hg pollution in the ambient air around AMM sites [6]. Workers involved in AMM in Hg mines were seriously exposed to Hg vapor through inhalation [59], resulting in highly elevated UHg. But for the local residents, dietary intake is the main pathway of IHg exposure [63], since the vegetables, rice, and meat samples are seriously polluted by IHg due to long-term Hg mining activities.

9.4.2 Health Risk Assessment

9.4.2.1 IHg Exposure

Occupational exposure to Hg vapor can have significant effects on the central nervous system. Exposure has resulted in erethism, irritability, excitability, excessive shyness, and insomnia as the principal features of a broad range of functional disturbances. With continuing exposure, a fine tremor develops, initially involving the hands and later spreading to the eyelids, lips, and tongue, causing violent muscular spasms in the most severe cases [68]. Gingivitis and typical dark lines on gums have been reported after high inhalation exposures [69]. Clinical symptoms include finger and eyelid tremor and gingivitis. Typical dark lines on gums were observed in six workers from the Wuchuan Hg mine [59] and three workers from the Tongren Hg mine [61]. These symptoms indicated that physical impairments had occurred and that workers were heavily exposed to Hg vapor during the process of cinnabar roasting.

Significant differences of urinary β2-microglobulin (β2-MG) concentrations were found between AMM workers in the Wuchuan Hg mine and the control group. β2-MG is a renal biomarker used to study human nephrotoxicity at an early stage. This biomarker is useful for assessing re-absorption function to indicate tubular injury. A significant correlation ($r = 0.85$, $p < 0.01$) was found between UHg and urinary β2-MG concentrations in the AMM workers, which confirmed that elevated urinary β2-MG concentrations in smelting workers does result from exposure to Hg vapor and supports the observation that Hg exposure results in serious adverse effects on the renal system in AMM workers.

For residents in the Wanshan Hg mine area, a significantly positive correlation is found between paired results for UHg concentrations and serum creatinine (SCr) ($p<0.05$). The SCr levels in the third (Q3) and fourth quartiles (Q4) of UHg were significantly higher than those from the first quartile (Q1) ($p < 0.05$ and $p < 0.01$, respectively). An increase of SCr levels was found in quartiles with the highest UHg concentrations, indicating that IHg exposure may result in renal impairment.

In conclusion, workers involved in AMM in Hg mines are extensively exposed to Hg vapor, which can cause neurologic damage and renal impairment. Local residents are also exposed to IHg in a certain level, which may have impact on renal function.

9.4.2.2 MeHg Exposure

The hair Hg threshold for onset of neurological symptoms in the human body is 50 μg/g [70], and the threshold for the onset of symptoms for MeHg is reported to be 10–14 μg/g in maternal hair [71]. The Joint Food and Agriculture Organization of the United Nations (FAO)/WHO Expert Committee on Food Additives [72] established a provisional tolerable

weekly intake (PTWI) for MeHg of 1.6 μg Hg/kg bw/week (0.23 μg Hg/kg bw/day, equivalent to a hair Hg concentration of about 2.3 μg/g) using an uncertainty factor 6.4, where bw = body weight. The United States Environmental Protection Agency [68] set the limit at 0.1 μg Hg/kg bw/day (equivalent to a hair Hg concentration of 1.0 μg/g) as a reference dose (RfD) using an uncertainty factor of 10.

The geometrical mean of hair MeHg concentrations in the population from Hg mines was 1.38 μg/g (CI 95% 0.50–5.06 μg/g). About 24.6% of inhabitants in the Hg mines were exposed to MeHg with hair MeHg concentrations exceeding 2.3 μg/g and 62.0% were exposed to MeHg with hair MeHg concentrations exceeding 1.0 μg/g. In addition to Wanshan, there are 11 other Hg mining and smelting areas in Guizhou [53], with total population approximately 320,000 [73]. Approximately 20,467 residents in Guizhou (0.06% of the total population) are exposed to MeHg with resulting hair MeHg concentrations exceeding 2.3 μg/g, and approximately 198,400 residents (0.57% of the total population) are exposed to MeHg with resulting hair MeHg concentrations exceeding 1.0 μg/g.

Generally, inhabitants in the Hg mines are exposed to MeHg to a certain level. However, they are not under serious health risk. Nevertheless, some female participants may exceed the tolerable intake levels of MeHg for pregnant women established by USEPA.

9.5 Summary

China is the third largest Hg-producing country after Spain and Italy. Although most large-scale Hg mining and retorting activities in China recently ceased operation, abandoned Hg mines are continuing to impact local environments through historic mine-wastes present in nearby adits, valleys, and riverbanks as well as through drainages. A large quantity of Hg compounds continues to be released into the surroundings via runoff, wind power, and precipitation, resulting in extensive Hg contamination of the environment. Results for human exposures from both IHg and MeHg suggest that potential human health risks exist in Hg mining areas. In the future, proper management and remediation of Hg contaminated environments in Hg mining areas in China are urgently needed.

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