



Review

Synthesis of current data for Hg in areas of geologic resource extraction contamination and aquatic systems in China

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ABSTRACT

China has become the largest contributor of anthropogenic atmospheric mercury (Hg) in the world owing to its fast growing economy and the largest of populations. Over the last two decades, Hg has become of increasing environmental concern in China and much has been published on its distribution, transportation, methylation, and bioaccumulation in aquatic systems and areas of geologic resource extraction contaminated sites, such as coal-fired power plants, non-ferrous smelters, Hg mining and retorting sites, Au amalgam, landfills, chemical plants, etc. Environmental compartments, like soil, water, air, and crop from areas of geologic resource extraction contamination, especially from Hg mining regions, exhibit elevated values of total-Hg and MMHg. Risk assessments indicate that the consumption of rice, which has a high bioaccumulation of MMHg, has become the dominant pathway of MMHg exposure of inhabitants living in Hg mining areas. Low concentrations less than 5 ng l⁻¹ in total-Hg can be observed in rivers from remote areas, however, high concentrations that reached 1600 ng l⁻¹ in total-Hg can be found in rivers from industrial and urban areas. The studies of hydropower reservoirs of southwest China indicated the old reservoirs act as net sinks for total-Hg and net sources of MMHg, while newly established ones act as net sinks for both total-Hg and MMHg, which is in sharp contrast to the evolution of biomethylation in reservoirs established in the boreal belt of North America and Eurasia. Fish from those reservoirs have relatively low levels of total-Hg, which do not exceed the maximum total-Hg limit of 0.5 mg kg⁻¹ recommended by WHO. Currently, however, there is still a large data gap regarding Hg even in the areas mentioned above in China, which results in poor understanding of its environmental biogeochemistry. Moreover, for a better understanding of human and environmental health effects caused by the fast growing economy, long-term Hg monitoring campaigns are urgently needed.

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1. Introduction

The toxic element mercury (Hg) is present in the environment as elemental Hg (Hg^0) and various inorganic and organic forms. Since Hg^0 is relatively chemical inert towards oxidation and moreover volatile, the species can undergo long-range transport in the atmosphere from the emission sources to remote areas before being deposited. Inorganic oxidised Hg exclusively exists in the divalent state ($Hg(II)$) as compounds predominantly associated with chlorine, sulphur, and hydroxyl ions. Organic Hg forms are more efficiently absorbed into the body than inorganic Hg and show high toxicities to the living (Clarkson and Magos, 2006). Mono-methylmercury (MMHg) species produced by aqueous biotic as well as abiotic processes from inorganic Hg is of the greatest concern to human health because of its bioaccumulation and high toxicity. Currently, the consumption of fish has been widely recognised as

the primary pathway of human Hg exposure except for the occupational populations (WHO, 1990).

Due to the fastest growing economy during the last two decades, China has become the world's largest Hg emitting nation with rapidly increasing Hg emissions to atmosphere. Emission inventories as well as modelling exercises confirm that non-ferrous metals smelting and coal combustion stand out as the corresponding predominant anthropogenic Hg source categories in China (Streets et al., 2005; Wu et al., 2006; Jaffe and Strode, 2008). In addition, metallic Hg production from cinnabar roasting and retorting and related gold extraction through amalgamation with continuity from ancient times are currently considered as another two important Hg releasing sources to the environment of China. On the other hand, China currently uses more Hg and its production than any other countries in the world, and on that account, releases more

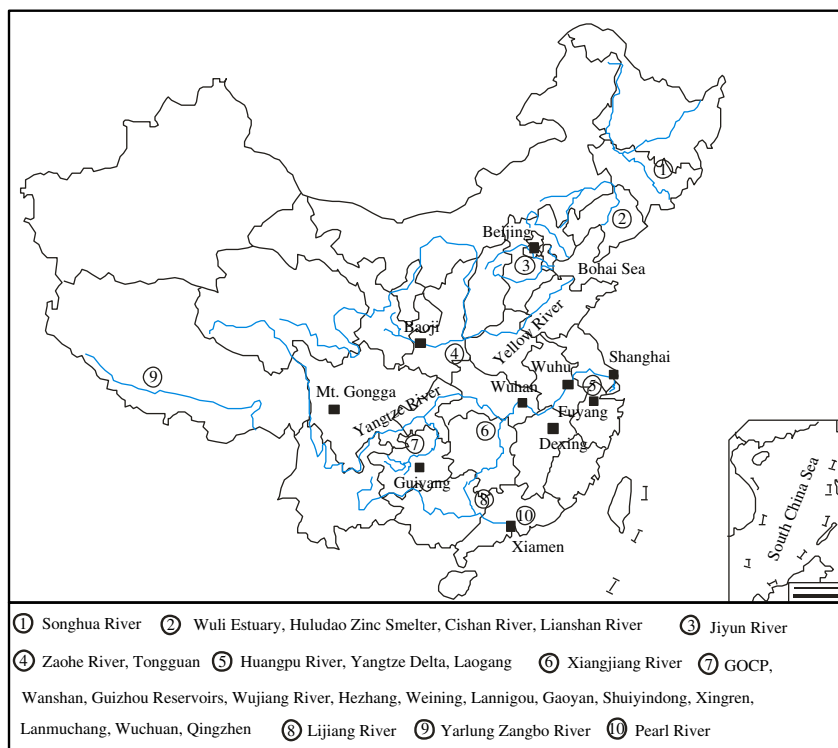


Fig. 1. Map of areas Hg studies reported in China.

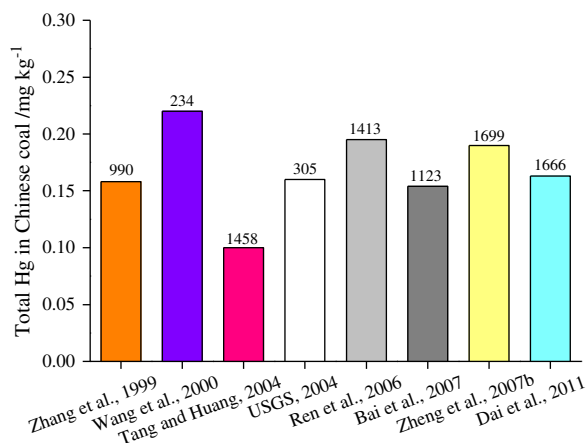


Fig. 2. Total-Hg concentrations in Chinese coals (the number of samples for the individual studies are indicated above the corresponding column).

Hg into the environment. For inventory concerning year 2004, the total supply of metallic Hg in China was 1400 t (Jiang et al., 2006a; NRDC, 2007), and among all the consumers, polyvinyl chloride (PVC) manufacturing utilising Hg compounds as catalyst, is the largest and was estimated to account for more than 40% of the Hg consumed (NRDC, 2007).

Anthropogenic released Hg contributes to environmental pollution and poses a long-term risk to wildlife and human beings when present as MMHg (WHO, 1990, 1991). The potential of Hg to undergo methylation in the ecosystems is of particular concern. However, a comprehensive understanding of chemical as well biological factors influencing the production and degradation of MMHg is lacking (Ullrich et al., 2001). Emerging as the world's largest consumer and emitter of Hg, China is thus expected to be continuously impacted by elevated Hg depositional flux over a long term in the future (Xiao et al., 1998; Tan et al., 2000; Fang et al., 2001; Jaffe and Strode, 2008; Fu et al., 2010a). To effectively manage the Hg pollution issues in the nation, a better understanding of temporal and spatial distribution of contaminants and biogeochemical process involving Hg in the environment are necessary. This review mainly focuses on giving an account of the current understanding of the distribution and speciation of Hg and related geochemical processes in areas influenced by geologic resource extraction contamination such as coal-fired power plant, non-ferrous smelter, Hg mining and retorting, Au extracting, chemical plants, landfills, etc., in which the aquatic systems in China is included (Fig. 1). This report also identifies areas in need of further research in such regions.

2. Mercury in areas of geologic resource extraction contamination

2.1. Coal-fired power plants

China is the world's largest coal consumer estimated to account globally for 46.9% (2.74 Gt/yr) of the total consumption (bp.com/statisticalreview). Therefore, coal combustion has increasingly become the predominant source category of anthropogenic Hg emissions to the environment in China.

2.1.1. Mercury in coal

Mercury exists in coal incorporated with other elements. The occurrence and concentration of Hg in coal from China has been the subject of numerous investigations (e.g. Zhang et al., 1999; Feng and Hong, 1999; Wang et al., 2000; Tang and Huang, 2004; USGS, 2004; Ren et al., 2006; Bai et al., 2007; Dai et al., 2011). Zhang et al. (1999) estimated an average of 0.158 mg kg⁻¹ Hg in Chinese coal calculated from 990

Table 1
Mercury concentration in coal from different provinces in China (mg kg⁻¹).

Province	Range	Mean	References
Anhui	0.060–0.79	0.22–0.26	Zheng et al., 2007a, 2008a, 2008b, 2008c; Wu et al., 2006; Wang et al., 2000
Beijing	0.10–0.54	0.34–0.44	Wu et al., 2006; Wang et al., 2000
Fujian	<0.02–0.14	0.07–0.08	USGS, 2004; Wu et al., 2006
Gansu	0.04–1.35	0.05	Wu et al., 2006; Ren et al., 2006
Guangdong	<0.02–	0.06–0.15	USGS, 2004; Wu et al., 2006; Ren et al., 2006
Guangxi	0.09–0.65	0.30–0.35	USGS, 2004; Wu et al., 2006
Guizhou	0.040–12	0.090–1.6	Dai et al., 2006; Wu et al., 2006; Zhang et al., 2004a; Feng et al., 2000
Hainan		0.15	Wu et al., 2006
Hebei	0.050–0.28	0.13–0.14	Wu et al., 2006; Wang et al., 2000
Heilongjiang	0.020–0.63	0.09–0.12	Wu et al., 2006; Wang et al., 2000
Henan	0.02–0.81	0.25–0.30	USGS, 2004; Wu et al., 2006; Wang et al., 2000
Hubei	0.12–0.24	0.16	USGS, 2004; Wu et al., 2006; Ren et al., 2006
Hunan	<0.02–0.32	0.08–0.14	USGS, 2004; Wu et al., 2006; Ren et al., 2006; Zheng et al., 2007b
Inner Mongolia	<0.02–2.1	0.15–1.5	USGS, 2004; Song et al., 2007; Wu et al., 2006; Wang et al., 2000; Zheng et al., 2007b; Ren et al., 2006
Jiangsu	0.11–0.60	0.16–0.34	USGS, 2004; Wu et al., 2006
Jiangxi	0.09–0.52	0.22–0.27	USGS, 2004; Wu et al., 2006; Ren et al., 2006; Zheng et al., 2007b
Jilin	0.050–1.6	0.07–0.33	USGS, 2004; Wu et al., 2006; Wang et al., 2000
Liaoning	0.020–4.8	0.14–0.17	Liu et al., 2009c; Wu et al., 2006; Wang et al., 2000; USGS, 2004; Ren et al., 2006
Ningxia	<0.02–1.2	0.20–0.44	USGS, 2004; Song et al., 2007; Wu et al., 2006; Ren et al., 2006
Qinghai		0.04–0.31	USGS, 2004; Wu et al., 2006; Ren et al., 2006
Shaanxi	<0.02–0.61	0.11–0.30	USGS, 2004; Wu et al., 2006; Ren et al., 2006; Wang et al., 2000
Shandong	0.03–0.97	0.13–0.38	USGS, 2004; Zheng et al., 2008a; Liu et al., 2007; Wu et al., 2006; Ren et al., 2006; Wang et al., 2000
Shanxi	<0.020–2.9	0.15–1.1	USGS, 2004; Ren et al., 2006; Song et al., 2007; Wu et al., 2006; Zhang et al., 2004c; Wang et al., 2000; Zheng et al., 2007b
Sichuan		0.11–0.35	USGS, 2004; Wu et al., 2006; Ren et al., 2006; Wang et al., 2000
Xinjiang	<0.02–0.28	0.02–0.24	USGS, 2004; Chen et al., 2007; Wu et al., 2006; Ren et al., 2006; Wang et al., 2000
Yunnan	0.03–0.42	0.14–0.32	USGS, 2004; Ren et al., 2006; Zheng et al., 2007b
Zhejiang		0.35–0.75	Wu et al., 2006; Ren et al., 2006

samples. Wang et al. (2000) reported a slightly high value of 0.22 mg kg⁻¹, with a range of 0.02–1.92 mg kg⁻¹. A collaborative study on Hg in Chinese coals between the U.S. Geological Survey (USGS) and the Institute of Geochemistry, Chinese Academy of Sciences (IGCAS) suggested an average Hg concentration of 0.16 mg kg⁻¹, within a range of <0.2–0.69 mg kg⁻¹ (USGS, 2004). Recently, Dai et al. (2011) reported an average Hg concentration of 0.163 mg kg⁻¹ for 1666 analysed coal samples. The above cited papers all told comprise 6199 samples with average total-Hg concentration reported in the separate studies for Chinese coal in the span from 0.10 to 0.22 mg kg⁻¹ (Fig. 2 and Table 1), in contrast to that of US coal (0.17 mg kg⁻¹) and coal globally (0.10 mg kg⁻¹) reported by Finkelman (1993) and Ketris and Yudovich (2009), respectively.

However, there is a large regional variability of the average Hg coal content within China (0.03 to 1.6 mg kg⁻¹) explained by the shifting geological premises. Following the provincial sub-division of Table 1, it is evident that Hg concentrations in coal samples from the provinces Guizhou, Inner Mongolia, and Shanxi vary

more widely. The investigation of Dai et al. (2006) of Hg content in coals from Xingren, Guizhou province revealed a maximum as high as 12.1 mg kg^{-1} . Samples collected at other locations of this province also exhibit high concentrations of Hg (Feng et al., 2000; Zhang et al., 2004b; Mukherjee et al., 2008). The high level of Hg in coals from Guizhou is consistent with its position within the circum-Pacific mercuri-ferrous belt. The province has up to 80% of the total cinnabar ore resources in China (Qiu et al., 2005).

The concentration of Hg in coal may depend on its density and geologic ages as well as other parameters, such as moisture, presence of sulphur, and carbon content (Mukherjee et al., 2008; Dai et al., 2011). The application of a representative average value for Hg concentration in coal requires careful consideration as our current knowledge on the chemistry of Hg in Chinese coal is still lacking.

2.1.2. Mercury in the surrounding environments

Mercury emitted from coal combustion mainly consists of gaseous elemental Hg (GEM), reactive gaseous Hg (RGM), and particulate Hg (HgP). RGM and a significant portion of HgP are susceptible to relatively rapid removal from the atmosphere, which occurs predominantly on a local scale (Bullock et al., 1998; Landis et al., 2004). An investigation on the spatial distribution of Hg content in soil surrounding a coal-fired power plant in Baoji, Shaanxi province revealed a divergent total-Hg distribution horizontally as well as vertically (range: 0.14 to 2.1 mg kg^{-1}). The mean Hg soil content of the study area (0.61 mg kg^{-1}) was significantly elevated compared to the provincial background level (Yang and Wang, 2008). Fang et al. (2010) reported that total-Hg concentration in soils impacted by a coal-fired power plant in Wuhu, Anhui province, ranged from 0.025 to 1.696 mg kg^{-1} , with an average of 0.212 mg kg^{-1} . The distribution characteristics of Hg in soil indicated that high levels were confined $\leq 1 \text{ km}$ distance downwind the plant, where soil-Hg was predominantly humic bound or contained in the residual fraction of the step-wise fractionation procedure employed.

Liu et al. (2011) conducted an investigation of the impact by a coal-fired power plant equipped with seawater flue gas desulphurisation system with respect to Hg pollution of the surrounding sea water (Xiamen region, Southern China). Elevated concentrations of aqueous Hg were found within 0.5 km of the waste drainage discharge outlets. Moreover, atmospheric GEM was comparatively elevated in this zone indicating that substantial air-sea evasion of Hg^0 occurred after the waste water was discharged into the sea body.

Quan et al. (2009) calculated Hg deposition from two cases of Hg speciation scenarios for coal-fired power plants in South-western China using a CMAQ-Hg model combined with meteorological observations. The results of Hg deposition showed RGM was the main component, which could contribute more than 90% of the total. High Hg emissions lead to high Hg deposition and total simulated Hg deposition due to coal combustion could reach as high as $2\text{--}6 \mu\text{g m}^{-2} \text{ month}^{-1}$. In the two cases of the total emitted Hg from coal combustion, approximately 40% was deposited in the studied regions and about 60% transported to other regions.

2.1.3. Mercury emission

Based on the estimated average Hg concentration in coal combined with total mass consumed, researchers have calculated Hg emission from coal combustion in China. Wang et al. (2000) estimated the total-Hg emission to the atmosphere from coal combustion in China was 213.8 t in 1995 whereas Streets et al. (2005) presented an estimate of 202.4 t for 1999. In the model presented by Wu et al. (2006) Hg emission from coal combustion was predicted to have increased from 202 t in 1995 to 257 t in 2003 at an average annual rate of 3.0%. In the study of Wu et al. (2006), the Chinese Hg emissions from coal combustion were speciated as 33% RGM and 10% HgP.

Several emission tests on-site measurement of Hg in process streams and flue gases of coal-fired power plants equipped with

various types of emission control devices have been conducted in China (Chen et al., 2007; Tang et al., 2007; Zhang et al., 2008; Wang et al., 2010). In Guizhou province, statistical data for coal consumption suggested that large-scale coal-fired power plants, industrial boilers, and domestic users consumed about 30%, 48%, and 22% of the total, respectively (Tang et al., 2007). Using a mass balance technique, Tang et al. (2007) estimated that Hg emission rates of industrial boilers without/with flue gas control device (FGD) systems were 66% and 26% of the total-Hg input, respectively, whereas Hg emission rates of coal-fired power plants with electro-static precipitation systems (ESPs) only were up to 85%, decreasing to 43% where FGD systems were in operation. According to the study of Tang et al. (2007), 56% of Hg in coal in Guizhou was released as Hg^0 , 33% as RGM, and 13% as HgP, which is comparable to the estimated distribution in China by Streets et al. (2005).

Wang et al. (2010) characterised Hg emission and its speciation/fractionation in six coal-fired power plants of China using the Ontario Hydro method. The Hg mass balances for the entire power plant varied from 87 to 116% of the input coal Hg and the average Hg removal efficiencies of three different air pollution control devices (APCDs) ESP, ESP + wet FGD, and ESP + dry FGD + fabric filters were 24%, 73% and 66%, respectively. The Hg emission factors of the six tested power plants exhibited a wide range from 11 to 130 mg Hg t^{-1} coal, implying the crucial role that the relevant APCDs play in curbing Hg emissions.

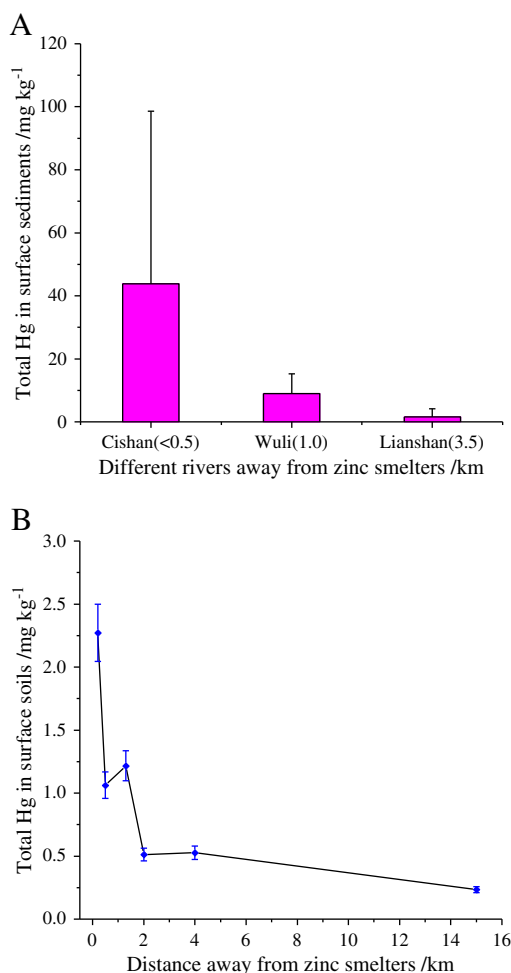


Fig. 3. Total-Hg concentrations in surface soil and sediments impacted by industrial zinc smelters (data from Zheng et al., 2008a, 2008b, 2008c, 2008d; Li et al., 2011).

2.2. Zinc smelters

China is abundant in zinc mines and the reserve of Zn ranks second in the world. The primary Zn production constitutes an important atmospheric Hg emission source since Hg is an important associate element in Zn ores. The rapid development of China's zinc industry, both as a producer and consumer, has led to this source category becoming the second largest in China concerning Hg emissions (Streets et al., 2005; Jaffe and Strode, 2008). In general, zinc smelting activities in China can be divided into hydrometallurgical (HP) and pyrometallurgical process (PP), where the latter is composed of many subtypes, such as retort Zn production, the imperial smelting process, the electric furnace, and the artisanal Zn smelting process (Li et al., 2010a). The Hg emissions largely depend on Hg contents in zinc ore and the specific air pollution control devices (APCDs) applied at the plant.

2.2.1. Mercury in zinc ore

A few data for Hg concentrations in zinc ores from Guizhou province of China are available. Feng et al. (2004a) reported that Hg concentrations in zinc ores from Hezhang County ranged from 35.4 to 157 mg kg⁻¹ for sulphide ores, and from 0.75 to 68.9 mg kg⁻¹ for oxide ores, respectively. Li et al. (2008a) found Hg concentrations in zinc ores from Weining County, an area adjacent to Hezhang, ranged from 1.1 to 87.1 mg kg⁻¹ for oxide ores, which was similar to the results found in Hezhang County. Elevated Hg concentrations in both sulphide Zn ores and oxide Zn ores occurring in the cited areas are presumably due to locations within the circum-Pacific Hg belt (Feng and Qiu, 2008).

2.2.2. Mercury in the surrounding environments

Total-Hg in surface water of streams impacted by the artisanal zinc smelters in Hezhang ranged from 12 to 691 ng l⁻¹ and reactive Hg from 0.5 to 4.9 ng l⁻¹, whereas total-Hg concentrations in the spring waters and well waters are much lower and were found in the range between 8.1 and 18 ng l⁻¹ and reactive Hg between 0.4 and 1.5 ng l⁻¹, respectively (Feng et al., 2004a). Total-Hg concentrations in surface water of streams impacted by similar activities in Weining were found to range from 95 to 278 ng l⁻¹, dissolved Hg from 87 to 117 ng l⁻¹, and total MMHg from 0.23 to 6.1 ng l⁻¹ and dissolved MMHg from 0.19 to 2.2 ng l⁻¹, respectively (Li et al., 2008a).

However, waters impacted by industrial zinc smelters may contain levels up to the µg l⁻¹ range. Dissolved Hg concentrations in a section of the Xiangjiang (Yangtze) River, Hunan province, Central China were, for example, between 0.02 and 1.12 µg l⁻¹ (Wang and Arne, 2003) while the Zn smelter waste waters discharged contained 1.5 to 2.3 µg l⁻¹ total-Hg (Li et al., 2011). Similarly, the concentrations of total-Hg in contaminated surface water by the Huludao zinc smelter were high, and ranged between 0.04 and 2.7 µg l⁻¹, and MMHg between 0.046 and 3.0 ng l⁻¹ (Wang et al., 2009).

Contaminated sediments from the Cishan River impacted by the Huludao zinc smelter had a high average concentration of 87 mg kg⁻¹ total-Hg. The River Wuli sediments were found to contain 13 mg kg⁻¹ total-Hg, and total-Hg concentrations in sediments of the River Lianshan ranged from 0.22 to 5.4 mg kg⁻¹ (Zheng et al., 2008d; Fig. 3A). Wang et al. (2009) reported concentration ranges of 0.5 to 64 mg kg⁻¹ total-Hg and of 0.12 to 35 µg kg⁻¹ MMHg in surface sediments from the same area.

Surface soils adjacent to the Huludao zinc smelter contained up to 15 mg kg⁻¹ in total-Hg (Zheng et al., 2007a). Compared to high levels of Hg caused by industrial zinc smelting, the concentrations of total-Hg were mostly less than 1.0 mg kg⁻¹ in surface soils from the artisanal zinc smelting activities in Hezhang and Weining areas (Feng et al., 2006a; Li et al., 2008a). Li et al. (2008a) reported

MMHg concentrations of 0.20 to 1.1 µg kg⁻¹ in contaminated soils near zinc smelters in Weining with a mean value of 0.52 µg kg⁻¹. The proportion of MMHg to total-Hg was less than 1% (range: 0.08–0.92%), which is in good agreement with the literature data (Horvat et al., 2003; Qiu et al., 2005).

Li et al. (2011) reported that Hg concentration in the soil impacted by industrial zinc smelters generally decreased with increasing distance from the smelting sites. Most of the Hg was accumulated in the upper most 20 cm depth of soil profile, and Total-Hg concentrations decreased sharply from 20 to 30 cm depth, and reached a relatively low level at depth of 40–50 cm (Fig. 3B). Total-Hg concentration in vertical soil profiles in Hezhang exhibited a similar trend and sharply decreased in depth, indicating that Hg in top soils was derived from atmospheric deposition (Feng et al., 2006a).

Zheng et al. (2007a) reported a concentration range of 35 to 3300 ng m⁻³ with an average of 1200 ng m⁻³ for total-Hg in air impacted by the plume from China's largest zinc smelter in Huludao City. GEM concentrations in ambient air of the Weining artisanal zinc smelting area measured by Lumex RA-915⁺ were highly elevated at sites adjacent to the smelting sites, ranging from 30 to 3814 ng m⁻³, and sharply declining away from the smelters into farmland (range 13–48 ng m⁻³) (Li et al., 2008). At a site 10 km distant from the smelters, however, GEM concentrations generally were less than 10 ng m⁻³, similar to the mean TGM/GEM concentrations obtained in urban areas, such as Guiyang, Beijing, Chongqing, and other areas in China (Liu et al., 2002; Feng et al., 2003a, 2004b; Li et al., 2005; Wang et al., 2006a).

2.2.3. Mercury emission

Hezhang and Weining are important artisanal zinc smelting areas in China. Feng et al. (2004a) estimated average Hg emission factors in Hezhang to be 155 g Hg t⁻¹ of Zn for sulphide ores, and to be 79 g Hg t⁻¹ of Zn for oxide ores, respectively. Li et al. (2008a) reported an average Hg emission factor in Weining of 75 g Hg t⁻¹ of Zn for oxide ores. The average Hg emission factor from sulphide ores was almost two times higher than that of oxide ores. For the industrial-scale Zn production plants, Li et al. (2010a) reported emission factors ranging from 5.7 to 112 g Hg t⁻¹ Zn produced, depending on Hg

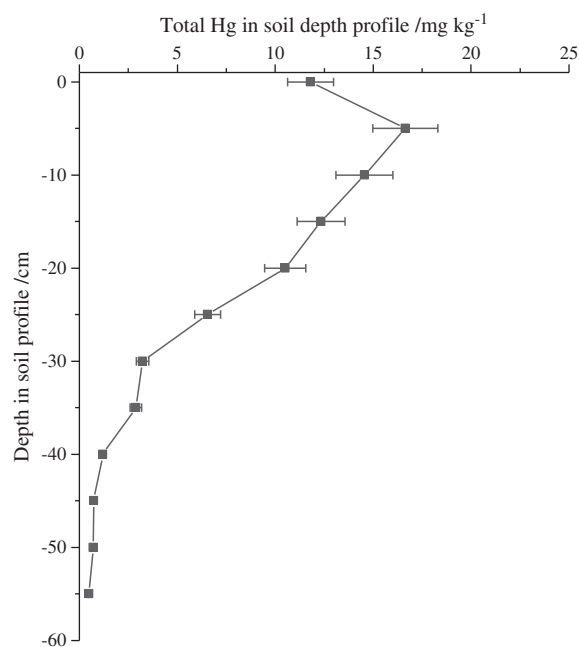


Fig. 4. Total-Hg in the soil depth profile from Hg mining areas (data from Qiu et al., 2006a).

concentrations in Zn ores as well as the APCDs employed. It is comparable to previous studies that reported emission factors from this sector ranging from 17 g Hg t⁻¹ of Zn produced (Hylander and Herbert, 2008) to 87 g Hg t⁻¹ Zn produced (Streets et al., 2005). Based on the Hg emission factors, Li et al. (2010a) estimated that Hg emission to the atmosphere from Zn production in China was 80.7 to 104.2 t year⁻¹ concerning the period 2002 to 2006. Currently, emission factors of 7.5–8.0 g Hg t⁻¹ of Zn for Europe, North America, and Australia, and of 20–25 g Hg t⁻¹ of Zn for Asia, Africa, and South America are widely accepted (Nriagu and Pacyna, 1988; Pirrone et al., 1996; Streets et al., 2005; Pacyna et al., 2006).

2.3. Mercury mining

Mercury mining and production sites currently attract environmental concern regarding Hg pollution in China owing to its long history of smelting activity. Many large Hg mining areas are located in Guizhou province of China, where the operation of the large-scale Hg mining companies (state owned) had been completely shut down in 2004. However, artisanal Hg mining is still operating at several abandoned Hg mining regions. As a legacy of the operations, the retorted cinnabar ores (termed calcines) of both the large-scale Hg mining and the artisanal Hg mining have not been properly disposed of but instead directly piled in valleys or flooded plains, where they continue to release Hg into the environment.

2.3.1. Mercury in calcines

Qiu et al. (2005, 2006a) reported an extremely high value of 4400 mg kg⁻¹ total-Hg in calcines from the Wanshan Hg mining areas. Contrary to the high total-Hg contents, MMHg concentrations in the calcines were low, ranging from 0.17 to 3.9 µg kg⁻¹. There is no significant correlation between MMHg and total-Hg contents present in calcines.

The process of Hg-retorting always leads to the accumulation of highly water-soluble secondary Hg phase forms in calcines (Kim et al., 2000). Though MMHg concentrations are much lower compared to those found at other Hg mining areas in the world, such as the McDermitt calcines in Nevada, USA (Gray et al., 2002) and the Almadén calcines in Spain (Gray et al., 2004), the abnormally high concentrations of inorganic Hg may suggest that the calcines act as significant Hg contamination sources to the surrounding ecosystems.

2.3.2. Mercury in the surrounding environments

In most of the abandoned Hg mining areas, high levels of Hg can be observed in surface waters derived from surrounding aquatic systems impacted by calcines. Total-Hg in surface water collected from Wanshan, Lanmuchang, and Wuchuan in Guizhou province generally varied in concentration range from 24.8 to 7020 ng l⁻¹ (Feng et al., 2003b; Qiu et al.,

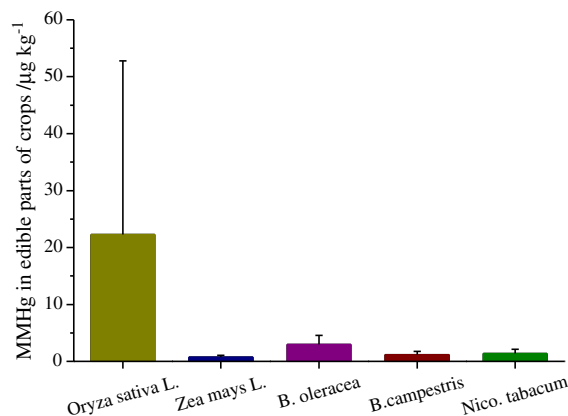


Fig. 5. Distribution of MMHg in edible parts of crop in Hg mining areas (data from Qiu et al., 2008).

2006a, 2006b, 2009; Li et al., 2008b, 2008c). Horvat et al. (2003) reported a high value of 10 580 ng l⁻¹ total-Hg in mining drainage emanating from calcines. A geochemical study also showed that drainage from calcines piles had high dissolved Hg concentrations of 300–1900 ng l⁻¹ with a pH value of 11–12 (Zhang et al., 2004a). A positive correlation between total-Hg and suspended substances was observed (Feng and Qiu, 2008), suggesting that Hg associated with particulate matters was an important factor for Hg migration from abandoned Hg mining sites.

Qiu et al. (2009) reported that MMHg in surface waters exhibited a wide concentration range of 0.3 to 25 ng l⁻¹. A significant positive correlation between total MMHg and dissolved MMHg in surface water was observed (Feng and Qiu, 2008), indicating that the dissolved fraction dominated MMHg levels in stream waters from Hg mining areas.

Total-Hg concentration in surface soils is largely heterogeneous and present in a wide range from the background levels to several hundred mg kg⁻¹. At Wanshan Hg mine, a total-Hg concentration up to 790 mg kg⁻¹ was measured (Qiu et al., 2005). As far as 24 km from the Wuchuan mine, soils alongside downstream river banks were still heavily contaminated with Hg (24 mg kg⁻¹) (Qiu et al., 2006a). Distribution of Hg in a soil depth-profile impacted by a long-term Hg deposition from a smelter showed the highest total-Hg concentrations in the top layer, ranging from 6.5 to 17 mg kg⁻¹ decreasing to a low level of 0.48 mg kg⁻¹ at 45 cm depth (Fig. 4). Elevated MMHg concentrations have also been observed in soils, ranging from 1.27 to 22.5 µg kg⁻¹ in the Wanshan Hg mining area (Horvat et al., 2003), and from 0.69 to 20 µg kg⁻¹ in the Wuchuan Hg mining area (Qiu et al., 2006a). No significant correlations between MMHg and total-Hg were found, however, ratios of MMHg to total-Hg were significantly lower in mine areas than in the background sites.

Pervious investigations have revealed that MMHg in paddies from Hg mining areas was higher than that in cornfields in the same areas (Qiu et al., 2005, 2006a). A further investigation indicated that the newly deposited Hg is more readily transformed to MMHg than Hg forms with an extended residence time in the paddy soil (Meng et al., 2011). Generally, the physico-chemical conditions present in the paddy environment facilitate Hg methylation due to the presence of a flora of sulphur-reducing bacteria, which probably contribute to the high levels of MMHg (Wind and Conrad, 1995; Stubner and Conrad, 1998; Hines et al., 1999). Moreover, in most Hg mining sites of China, GEM in ambient air can attain largely elevated concentrations, resulting in significant deposition fluxes (Wang et al., 2005, 2007a, 2007b). The issue of the promotional effect of newly deposited Hg on the methylation process in the paddy environment needs to be further scrutinised in future studies.

2.3.3. Mercury in crop

Table 2 lists results for some studies concerning Hg (total-Hg and MMHg) concentrations in crops from the Hg mining areas of

Table 2
Total-Hg and MMHg concentration in plant samples collected from Hg mining areas in China.

Sample	THg/mg kg ⁻¹		MMHg/µg kg ⁻¹		References
	Range	Mean	Range	Mean	
Rice	0.009–1.1	0.13–0.23	1.6–174	11–39	Horvat et al., 2003; Cheng et al., 2006; Qiu et al., 2006a, 2008; Feng et al., 2008
Corn	0.089–0.57	0.065–0.20	0.28–13	0.71	Qiu et al., 2006a, 2006b; Cheng et al., 2006
Cabbage	0.12–18	2.5	0.65–5.5	3.2	Qiu et al., 2006a, 2006b
Cole	0.47–0.69	0.57	0.57–0.83	0.71	Qiu et al., 2006a, 2006b
Tobacco	0.20–0.76	0.48	0.83–1.9	1.4	Qiu et al., 2006a, 2006b
Potato		0.018			Cheng et al., 2006
Capsicum		0.18			Cheng et al., 2006

China. Outstanding is the range of up to 18 mg kg^{-1} total-Hg occurring in green cabbages (*B. oleracea*) grown in Hg-contaminated soils in Lanmuchang (Qiu et al., 2006b), but other crops, such as corn, rape, tobacco grown in Hg mining sites also exhibited high content of total-Hg (Qiu et al., 2006a, 2008).

Although inorganic Hg species occur predominantly in Hg-enriched plants, high MMHg concentrations in rice (*Oryza sativa* L.) grown at Hg mining areas have been reported as well (Table 2). Data for MMHg showed that rice grown from Hg mining areas could assimilate and accumulate MMHg in its edible portion, which was found to be 10–100 times higher than in other crop plants (Fig. 5). Horvat et al. (2003) and Qiu et al. (2008) found high MMHg concentrations in rice seeds (edible parts) up to 0.14 mg kg^{-1} in Wanshan, and 0.17 mg kg^{-1} in Tongren, respectively. Meng et al. (2010a) conducted an investigation on distribution patterns of inorganic Hg and MMHg in tissues of rice plants and observed that rice seed has the highest ability to accumulate MMHg compared to the other tissues. Recently, a further study including sampling during the full rice growing season indicated that MMHg in paddy soils was initially absorbed by roots and then translocated into the above-ground parts, and eventually most of this MMHg was transferred to seed during the ripening period (Meng et al., 2011).

Available data on MMHg in rice reveal high concentrations in rice from Hg mining areas as well as from areas in the vicinity of chemical plants (cf. Section 2.4.4). Li et al. (2010c) investigated the speciation of MMHg in rice from Hg mining areas and observed that in rice grain MMHg is bound almost exclusively to the amino acid L-cysteinate ($\text{CH}_3\text{-HgCys}$). They also found that cooking does not change total MMHg in rice, but the $\text{CH}_3\text{-HgCys}$ is almost quantitatively converted into MMHg complexes of other ligands. Owing to the position of rice as the staple food for a large group of populations, the accumulation of MMHg in rice not only grown in Hg mining areas but also in impacted industrial regions should be investigated. This also applies for the mechanism of MMHg uptake into seeds, which is not yet clear.

2.3.4. Mercury emissions

Wang et al. (2005, 2007a, 2007b) investigated Hg emissions in the Hg mining areas of Guizhou (Lanmuchang, Wuchuan, and Wanshan) using dynamic flux chamber (DFC) technique. A correlation was found between the Hg soil content and the magnitude of Hg release to atmosphere. An average Hg emission flux of $19.9 \text{ mg m}^{-2} \text{ yr}^{-1}$ from surface soil was obtained at the Lanmuchang Hg mine, and within a total area of $\sim 2.9 \text{ km}^2$, an annual Hg emission rate was estimated to be $\sim 3.54 \text{ kg Hg}$, indicating a strong aerial Hg emission source to ambient air (Wang et al., 2005). Plot measurements in the Wanshan Hg mining area yielded average Hg emission fluxes in the range from 1.4 to $243.7 \text{ mg m}^{-2} \text{ yr}^{-1}$ while an average Hg flux estimate for the Wuchuan Hg mining area was significantly lower at $1.23 \text{ mg m}^{-2} \text{ yr}^{-1}$ (Wang et al., 2007a, 2007b).

Compared to diffusive Hg degassing from abandoned Hg mining areas, Hg emissions measured from artisanal retorting activities are huge due to its primitive character without any environmental protection devices employed. Tan et al. (1997) estimated that between 1978 and 1997, approximately 260 kg of Hg were lost to the environment of the Wuchuan Hg mining area during artisanal retorting. Using a mass balance technique, Li et al. (2006) estimated Hg emission rates from artisanal Hg retorting in the Wuchuan-Tongren area of NE Guizhou. Recoveries ranged from 78.4 to 93.6% depending on the retorting technique applied yielding a Hg emission estimate of 3.7–9.6 t per year. The investigation of Li et al. (2006, 2009a) highlights that indigenous Hg retorting activities currently represent the largest source of Hg to the atmosphere in related Hg mining areas.

2.3.5. Health risk assessment

Human health risk assessment studies on subjects of occupational Hg exposure in the Hg mining areas have been conducted. In Wuchuan, the average hair total-Hg and MMHg content of smelting workers was 69.3 and 2.32 mg kg^{-1} , respectively, being significantly

higher than that of subjects in a control group (0.78 mg kg^{-1} and 0.65 mg kg^{-1} , respectively) (Li et al., 2008d). Urinary Hg levels ranged from 22.6 to 4577 mg kg^{-1} creatinine for the exposed workers and from 1.0 to 17.4 mg kg^{-1} creatinine for the subjects in the control group (Sakamoto et al., 2007; Li et al., 2008d). Total-Hg concentrations in urine and hair from the exposed group displayed a significant correlation ($r=0.62$), indicating adhesion of Hg vapour to hair (Sakamoto et al., 2007). In Wanshan, Feng et al. (2008) reported that concentrations of total-Hg and MMHg in hair samples collected from the residents ranged from 0.6 to 58.5 mg kg^{-1} and 0.2 to 5.6 mg kg^{-1} , respectively. The hair Hg levels in subjects from a control site were much lower, for total-Hg and MMHg in the range from 0.32 to 1.7 mg kg^{-1} and from 0.26 to 1.1 mg kg^{-1} , respectively.

As MMHg tends to accumulate in rice, which is a staple food for residents, consumption of MMHg-contaminated rice may represent a social and economic problem. Rats kept on a diet of rice from Wanshan for 20 days revealed an inducible lipid peroxidation in brain, liver, and kidney tissue being associated with an increase of c-jun mRNA (Cheng et al., 2005, 2006). A study of 98 persons living in Wanshan Hg mining area showed a significant correlation ($r=0.65$, $p<0.01$) between hair MMHg concentrations and MeHg exposure through daily rice consumption (Feng et al., 2008). A rough calculation for adult daily intake of MMHg via rice consumption indicated an average value of $0.26 \mu\text{g kg}^{-1}$ of body weight per day, which is slightly higher than the new dietary reference dose (RfD) of $0.23 \mu\text{g kg}^{-1}$ of body weight per day for MMHg recommended by United Nations Committee (WHO, 2003; Qiu et al., 2008). Zhang et al. (2010) recently reported that the probable daily intake (PDI) of MMHg for an adult population ranged from 0.015 to $0.45 \mu\text{g kg}^{-1}$ per day in Wanshan. The results indicated that rice is an important MMHg exposure route for residents in the Wanshan Hg mining area and the main human exposure to MMHg via food consumption is through rice rather than fish.

2.4. Other hotspot areas

2.4.1. Gold mining

Gold mining activities play an important role in China's economic progress (Lin et al., 1997), but give rise to serious environmental problems. A few studies have been conducted concerning the environmental effects resulting from gold extraction using Hg amalgamation. Lin et al. (1997) reported elevated total-Hg concentrations (range 500 – $1000 \mu\text{g l}^{-1}$) in the wastewater discharged without treatment from gold mining in Dexing, Jiangxi province. Recently, Hg species in water samples collected in the gold mining area in Tongguan, China were reported (Dai et al., 2003; Feng et al., 2006b) and it was observed that typically $\gg 99\%$ of Hg in surface water was

Table 3
Total-Hg concentration in MSW and covered soils of landfills in China.

Location	MSW/ mg kg^{-1}		Soil/ mg kg^{-1}		References
	Range	Mean	Range	Mean	
Guiyang, Guizhou	0.17–46	0.32–1.9	0.13–6.5	0.18–3.2	Feng et al., 2004c; Tang et al., 2004; Li et al., 2010a
Beijing	0.23–1.0	0.60			Ren et al., 2001
Laogang, Shanghai			0.018–0.26	0.071	Tang et al., 2003
Three Gorges area, Chongqing	0.09–5.5				Wang and Chen, 2002; Wang et al., 2006b
Yanan, Shaanxi	0.005–0.54	0.20			Liu et al., 2005
Wuhan, Hubei	0.24–1.3	0.61	0.037–0.099	0.058	Li et al., 2010b

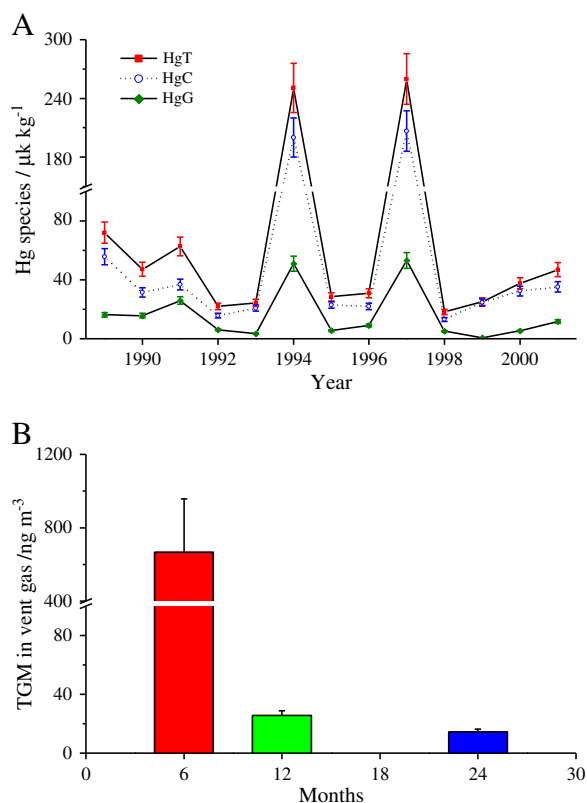


Fig. 6. Concentrations of total-Hg (HgT), matrix combined Hg (HgC), and gaseous Hg (HgG) in topsoil and trends of TGM in vent gas of landfills (data from Tang et al., 2003; Ding et al., 2005a, 2007; Tang et al., 2004).

bound to particulate matter. Dai et al. (2003) reported that 1.9–62.3% of dissolved Hg occurred as reactive Hg species in the study area. Total-Hg concentrations in sediments varied widely from 0.90 to 1200 mg kg⁻¹ (Feng et al., 2006b).

The study of Lin et al. (1997) indicated serious environmental Hg pollution in the gold producing area, where total-Hg concentrations in solid tailings and soils near the workshops were highly elevated (up to 100–300 mg kg⁻¹ and 1100 mg kg⁻¹, respectively). Soil samples collected in the vicinity of gold mills in the Tongguan gold mining area were also significantly contaminated (concentration range of total-Hg: 0.9 to 76 mg kg⁻¹) (Feng et al., 2006b). Soil samples from Hg-contaminated croplands in the same region have an average total-Hg concentration of 2.0 mg kg⁻¹ (Xu et al., 2006). Total-Hg in soil from the Lannigou carlin gold mine, Guizhou province, was reported to fall in the range between 1.6 and 37 mg kg⁻¹ with an average of 17 mg kg⁻¹ (Li and Zhang, 2009). Mean concentration of total-Hg (7.0 mg kg⁻¹) in soil samples from the Shuiyindong gold mining region, Guizhou province was also elevated (Wang and Zhang, 2009).

Airborne Hg concentrations inside workshops of a small scale gold amalgamation mining facility ranged from 2.0 to 2.8 mg m⁻³ close to Hg⁰ vapour saturation (Lin et al., 1997), which significantly exceeds the upper limit (0.01 mg m⁻³) for occupational exposure by China's national standard. TGM concentrations in ambient air of the Tongguan gold mining area were significantly elevated compared to the regional background, with a maximum of 18 µg m⁻³ observed at a gold mill (Feng et al., 2006b).

Total-Hg concentrations in vegetable and wheat samples in Tongguan fall in the range 42–640 µg kg⁻¹, significantly exceeding the Chinese guidance limit of 10 µg kg⁻¹ for vegetables and of 20 µg kg⁻¹ for food-stuffs other than fish. MMHg was not significantly elevated (0.2–7.7 µg kg⁻¹) in the crops sampled in this study (Feng et al., 2006b).

Bryophytes inhabiting two gold mining area of Guizhou province were found to contain high levels of total-Hg (Lannigou area: 1.7–7.8 mg kg⁻¹ (Li and Zhang, 2009), Shuiyindong region: 1.1–3.5 mg kg⁻¹ (Wang and Zhang, 2009)).

2.4.2. Landfills

Municipal solid waste (MSW) usually contains significant amounts of Hg due to the inclusion of Hg-bearing items, such as fluorescent lamps, thermometers, batteries, electrical and electronic equipments. In developed countries, the incineration of MSW (MSWI) forms an important category of sources contributing to atmospheric Hg emissions (Pirrone et al., 1996). In China, however, the major way to dispose of MSW (up to 90%) is through landfills.

Analytical data on the total-Hg content in MSW samples collected nationwide in China show a broad distribution over three orders of magnitude (0.005 to 5.5 mg kg⁻¹) with a single sample from Gaoyan, Guizhou containing 46 mg kg⁻¹ as an outlier (Table 3). The data compared favourably with surveys conducted in other countries, such as that of Alachua landfill in California, USA where MSW contained 0.033–17 mg total-Hg kg⁻¹ (Earle et al., 1999). In the study of Li et al. (2010b) including five landfills from the inland cities of Guiyang and Wuhan, the mean total-Hg concentration in MSW varied for the individual sites between 0.20 and 1.9 mg kg⁻¹.

The total-Hg concentrations in soil covering the Guiyang landfills showed an elevated range of 0.5–6.5 mg kg⁻¹ compared to the soil (total-Hg: 0.018–1.0 mg kg⁻¹) from the other investigated landfills (Tang et al., 2003; Li et al., 2010b). Total-Hg, gaseous Hg, and matrix combined Hg concentrations in topsoil of the Laogang landfill, Shanghai were found to be significantly positively correlated with each other, but no correlation between Hg concentrations and filling periods was obtained (Fig. 6A; Ding et al., 2005a, 2007).

Feng et al. (2004c) and Tang et al. (2004) investigated volatile Hg species (GEM, MMHg species, dimethyl Hg) degassed through vents from a landfill in Guiyang. A decreasing trend of the TGM concentrations in the vent gas was observed with increasing age of the landfill (Fig. 6B). This is evident also concerning the organomercurial species present in gas, where the MMHg concentration in 6-month and 24-month aged vent gas was 2.1 ± 1.8 ng m⁻³ and 0.18 ± 0.06 ng m⁻³, respectively. As landfill gases in China are rarely utilised or promptly flared, there is an ecological risk associated with a direct release of highly toxic dimethyl Hg and MMHg species from landfills.

Li et al. (2010b) investigated Hg emissions from landfills in Guiyang using DFC technique. GEM concentrations in ambient air ranged between 1.6 and 474 ng m⁻³. The mean Hg flux was 502 ng m⁻² h⁻¹ (range: 286–5610 ng m⁻² h⁻¹) at landfills without soil covering and was 55.2 ng m⁻² h⁻¹ (range: –72.5–309 ng m⁻² h⁻¹) at landfills with soil covering. At closed landfill sites, Hg flux ranged from 53.6 to 559 ng m⁻² h⁻¹, which was comparable to that observed from active landfills. According to measured Hg flux, approximately 600 kg Hg was emitted into atmosphere from landfills of China in 2004.

In the future, it is inevitable that more and more incineration facilities will be constructed in China to dispose of MSW instead of landfills. MSWIs are predicted to play an increasingly important role in municipal solid waste management in the next few decades in China, and therefore much greater effort in evaluating the environmental impact by the MSWI is required.

2.4.3. Copper smelter

Yin et al. (2009) conducted an investigation on Hg contamination in a secondary Cu manufacturing industrial region, Fuyang, Zhejiang province. Elevated total-Hg concentrations ranging from 0.033 to 15 mg kg⁻¹ were observed in the topsoil (0–15 cm) in the vicinity of Cu smelters. The corresponding total-Hg content was found to depend on the land-use type, with the mean Hg levels in uncultivated soil higher than that in cultivated soil, which has been attributed to

Table 4
Speciated Hg concentration in aquatic systems of China.

Locations		Water				Sediments				Pore water				References	
		HgT/ng l ⁻¹		MMHg/ng l ⁻¹		HgT/mg kg ⁻¹		MMHg/μg kg ⁻¹		HgT/ng l ⁻¹		MMHg/ng l ⁻¹			
		Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean		
River	Haihe					0.055–0.174	0.113	0.7–1.5	1.07					Shi et al., 2005	
	Huangpu	120–1580	440			0.07–0.39	0.20			320–2810	1310			Ding et al., 2005b	
	Lijiang	51–270	120			0.083–0.46	0.20							Qian et al., 2010	
	Mt. Gongga	1.5–7.3	3.6											Fu et al., 2010a	
	Wujiang	2.6–330	17–66	0.070–0.70	0.14–0.17									Jiang et al., 2003, 2004; Guo et al., 2008	
	Yarlung Zangbo	1.46–4.99	2.8	0.06–0.29	0.12									Zheng et al., 2010	
	Zaohe	99.2–992.7	453.8			0.032–0.38								Jin et al., 2008	
Lake and Reservoir	Aha	2.14–16.6	8.90	0.03–0.90	0.34	0.16–0.252	0.209	0.22–7.21	1.76	2.7–92.1	6.3–34.1	0.06–4.2	0.62–1.11	Bai et al., 2011; Feng et al., 2011	
	Baihua	20–336	76			0.26–39	13	0.5–27.5	6.1	6.1–5860	815	0.3–15	4.8	Feng et al., 2003c, 2004d; Hou et al., 2004; Yan et al., 2005, 2008	
	Caohai	1.7–9.0	5.3	0.11–0.67	0.25	0.76–1.01	0.876	0.11–3.9	1.1	8.6–40	18	0.11–4.9	1.8	Qian et al., 2008, 2009	
	Dongfeng	1.3–39	9.1	0.25–1.6	0.65	9.0–61	0.14–0.23	0.17	0.–2.8		5.0–30.0		0.35–5.0	2.1	Jiang et al., 2006b; Feng et al., 2009a, 2009b
	Hongfeng	2.5–14	6.9	0.053–0.33		24–110		0.39		2.4–3.4	14–23		0.76–1.6	He et al., 2006, 2008a, 2008b, 2008c	
	Hongjiadu	0.3–6.6	1.4	0.05–0.17	0.11									Yao et al., 2011	
	Puding	1.0–11.7	2.7	0.06–0.51	0.32					2.6–11.5	5.0	0.06–1.2	0.47	Zhang et al., 2009b; Liu et al., 2009a	
	Suofengying	0.4–4.9	1.3	0.03–0.22	0.11									Yao et al., 2011	
Wujiangdu	6.1–26	10	0.47–1.8	0.82	17–372	0.14–0.37	0.25	0.–7.9				0.12–3.1	0.81	Jiang et al., 2006b; Feng et al., 2009a, 2009b; Meng et al., 2010b	
Coastal and Estuary	Yinzidu	0.40–1.9	1.0	0.028–0.44										Meng et al., 2010b	
	Pearl River Estuary					0.0015–0.20								Shi et al., 2010	
	Yellow River Estuary					0.012–0.12								Liu et al., 2009b	
	Wuli Estuary	84–2700	690	0.046–0.064		44–64	51	8.0–26						Wang et al., 2009	
	Jinzhou Bay	39–430		0.046–0.28	0.14	0.80–25	6.8	0.37–5.2	0.057					Wang et al., 2009	
	Laizhou Bay					0.018–0.38								Liu et al., 2009b	
	East China Sea	1.51–92.88				0.055–0.201		0.017–0.256						Fang and Chen, 2010; Liang et al., 2010	
South China Sea	0.80–2.3	1.2	0.050–0.22	0.12	20.4–80.1	0.020–0.13		0.010–0.053					Kannan and Falandysz, 1998; Fu et al., 2010b		

ploughing homogeneities, plants scavenged, and adjacency to the smelters. Within an area of 10.9 km², a total amount of approximately 365–561 kg Hg emitted from the smelters was estimated to accumulate in topsoil in the region. The estimated Hg emission factors varied in the range 98.9–155.4 g Hg t⁻¹ of Cu produced by the secondary smelting, which is about 17–27 times higher than the emission factor of 5.81 g Hg t⁻¹ of Cu used to estimate the global Hg emission from copper smelting sector (Yin et al., 2009).

2.4.4. Chemical plants

Prior to the 1990s, the Songhua and Jiyun Rivers were seriously contaminated by Hg by chemical plants (cf. Section 3.1.2). The Guizhou organic chemical plant (GOCP) is unique in China because it uses metallic

Hg as a catalyst to produce acetic acid. The factory located in Qingzhen County, Guizhou province, has utilised Hg for that purpose for nearly 30 years. Yasuda et al. (2004) demonstrated that paddy fields downstream of the plant were a receptor for waste water and were seriously contaminated with Hg. The mean total-Hg concentrations in paddy soil reached 16 ± 43 mg kg⁻¹, significantly elevated compared to that of a control site (0.11 ± 0.05 mg kg⁻¹) nearby. Horvat et al. (2003) found that water, sediments and soil downstream the GOCP plant were active sites for transformation of inorganic Hg into organic Hg species. Surface waters heavily impacted by wastewaters from the plant were determined to contain up to 1830 ng l⁻¹ total-Hg. Total-Hg concentration in rice grains of the area of up to 88 μg kg⁻¹, of which 32.9 to 52.8% was in the form of MMHg. The results of Horvat et al. (2003) suggest

the potential of rice grown not only in Hg mining areas but also in Hg contaminated areas in general to be contaminated with MMHg. The number of studies on this topic is however limited and further study is needed to verify this hypothesis.

3. Mercury in the aquatic environment

Prior to the 1990s, most of the reported data concerning Hg in Chinese aquatic systems were focused on the Songhua River and the Jiyun River in Northeast and East China, respectively, which were heavily contaminated with Hg from the release of wastewater from organic chemical plants. The related literature mainly described the distribution of Hg species and its transport in both aquatic systems (e.g. Zhang et al., 1981; Lin et al., 1983; Wang et al., 1985; Zhang et al., 1994; Liu et al., 1997). The following discussion is mainly focused on the recent Hg data for other aquatic systems in China. A part of data from those investigations is listed in Table 4.

3.1. Mercury in water

3.1.1. Rivers

Studies have indicated that uncontaminated fresh waters generally contain $<5 \text{ ng l}^{-1}$ total-Hg (e.g. Bloom, 1989), which appears to apply for total-Hg in rivers from remote areas in China. Zheng et al. (2010) reported total-Hg concentrations in a range of 0.46 to 4.99 ng l^{-1} for the Yarlung Zangbo River, Tibet. The corresponding range of reactive Hg was 0.10 – 0.36 ng l^{-1} , while up to 70% of total-Hg was controlled by the particulate Hg. In the Mt. Gongga area, a remote region in Southwest China, Fu et al. (2010a) observed that the average total-Hg concentration in freshwaters of glacial streams was $3.5 \pm 0.9 \text{ ng l}^{-1}$.

Compared to the data from remote areas, Hg concentrations in surface water from many inland parts of China showed much elevated levels. Jiang et al. (2004) reported that total-Hg concentrations in Wujiang River, the largest tributary of the upper Yangtze River, varied from 2.6 to 329.6 ng l^{-1} , and particulate Hg was the predominant form. The elevated Hg concentrations in Wujiang River are probably to a large extent derived from the naturally elevated Hg content in the bedrock of the watershed located in a mercuriferous belt (Feng

and Qiu, 2008). Qian et al. (2010) observed total-Hg concentrations in the Lijiang River system, a scenic destination heavily frequented by tourists in South China, varied in the range 51 – 270 ng l^{-1} and 110 – 260 ng l^{-1} for the mainstream and tributaries, respectively.

In industrial and urban settings, riverine surface waters have occasionally been found to contain highly elevated total-Hg concentrations up to a $\mu\text{g l}^{-1}$ range. Ding et al. (2005b) reported a maximum of $1.6 \mu\text{g l}^{-1}$ that total-Hg concentration consisting of $>90\%$ as dissolved form in the section of the Huangpu River located in Shanghai City, East China. The Zaohe River, a tributary of Weihe River, also exhibited elevated total-Hg concentrations between 99.2 and 992.7 ng l^{-1} when passing through Xi'an City, Northwest China (Jin et al., 2008). High levels of Hg in rivers of the industrial and urban areas indicate a negative impact of the rapidly developing economy in China.

3.1.2. Lakes and reservoirs

Most studied lakes and reservoirs in terms of Hg distribution were located in Guizhou province along the Wujiang River. The nine reservoirs investigated (Baihua, Hongfeng, Aha, Wujiangdu, Yinzidu, Hongjiadu, Suofengying, Puding, and Dongfeng) represented a range of ages since construction and were to various degrees influenced by the local pollution sources. Combined published data from those reservoirs are listed in Table 4.

Experimental data confirmed that reservoirs without discernible discharges from industrial activities appear to have a comparatively low level of total-Hg and MMHg in waters (cf. Wujiangdu and Yinzidu Reservoir; Feng et al., 2009a, 2009b; Meng et al., 2010b). Furthermore, total-Hg concentrations in the Hongfeng Reservoir impacted by a coal-fired power plant were elevated and slightly increased with depth in water columns (He et al., 2008a, 2008b). The Baihua Reservoir, which has a recent history of Hg pollution caused by waste water discharges from the Guizhou organic chemical plant (GOCP), utilising metallic Hg as a catalyst to produce acetic acid, exhibited high levels of total-Hg in the water body and in underlying sediments (Feng et al., 2003c, 2004d; Hou et al., 2004; Yan et al., 2005). In a comparison between an up to 30 years old reservoir (Wujiangdu Reservoir) and newly constructed ones (Hongjiadu and Suofengying Reservoirs), the latter category exhibits slightly lower MMHg concentrations (Yao et al., 2011).

He et al. (2008a) conducted a long-term monitoring study of the Hg load in inflows and the outflow of the Hongfeng Reservoir and found a good comparison concerning the MMHg concentration in the flows with the exception for the summer period when the MMHg level in the outflow was up to 6 times higher than that in inflows (Fig. 7A and B). Feng et al. (2009a) reported that mean concentrations of MMHg in outflows of the Dongfeng and Wujiangdu reservoirs were 0.77 ng l^{-1} and 0.89 ng l^{-1} , respectively. Guo et al. (2008) reported total MMHg concentrations of inflows and outflows in those cascade reservoirs, ranging from 0.07 to 0.70 ng l^{-1} and from 0.10 to 0.34 ng l^{-1} , respectively.

The studies of reservoirs in the Wujiang River system indicated that the old reservoirs acted as net sinks for total-Hg but net sources of MMHg, while newly established ones acted as net sinks for both total-Hg and MMHg. Furthermore, the older reservoirs had a considerably higher MMHg production than the younger ones, and methylation usually occurs at the bottom of the water column or/and water-sediment interface. This is in sharp contrast to the evolution of Hg methylation in reservoirs established in the boreal belt of North America and Eurasia, where an initial high MMHg production was observed followed by a decreasing trend. This was attributed to an initially high availability of fresh organic matter and a subsequent decline in the availability of organic material for the methylating bacteria (Lucotte et al., 1999; St. Louis et al., 2004). In Guizhou, the area flooded after establishment of a new reservoir generally consisted of farmlands with a meagre organic matter content (1.9% to 4.1% for the Hongjiadu and Suofengying reservoirs (Yao et al. (2011)).

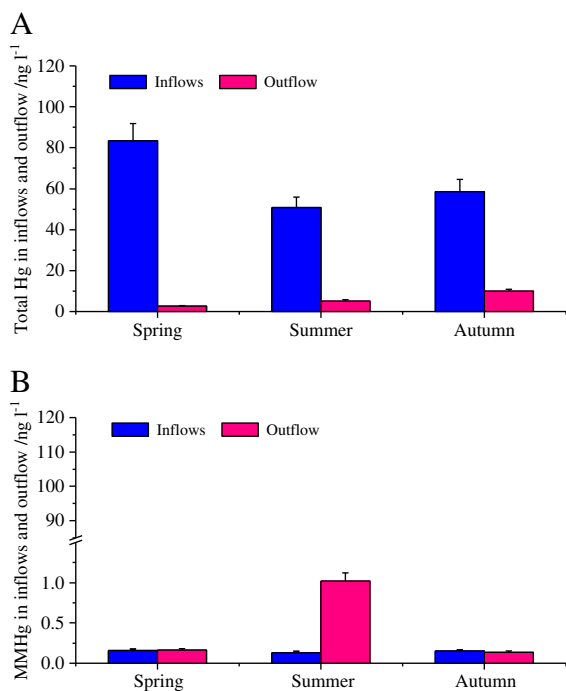


Fig. 7. Total-Hg and MMHg concentrations in inflows and outflows for a number of reservoirs in Guizhou province (data from He et al., 2008a; Jiang et al., 2006a, 2006b).

Hence, the limited access to fresh organic material may curb the establishment of active environments for biomethylation. However, to an increasing extent the reservoirs are affected by human activities, such as the cage aquaculture, that significantly add to organic matter in the sediment over time, feasibly stimulating the process of methylation. Feng et al. (2009a, 2009b) and Meng et al. (2010b) demonstrated that the primary productivity and also the water residence time are important factors that control the potential of net Hg methylation in the Wujiang River reservoirs.

Compared to lakes and reservoirs located at higher latitudes, the premises and evolution of methylation in Chinese hydropower reservoirs appear different. Since China is in the frontline concerning the construction of new reservoirs for hydro-electric reservoir in the world, increasing net Hg methylation due to high levels of primary production driven by eutrophication may become an emerging environmental issue (Larssen, 2010).

3.1.3. Coastal areas and estuaries

The coastal areas of China are characterised by intensive industrial, commercial, and recreational activities and are currently the world's leading hub for low-cost manufacturing. Related operations are considered to be potential sources of Hg release to the coastal zone environment. Wang et al. (2009) reported an elevated total-Hg concentration of 2700 ng l⁻¹ in surface water samples collected in the Wuli Estuary of Jinzhou Bay (northwestern coast of the Bohai Sea), which is impacted by a Zn smelter as well as a chlor-alkali plant. Fortunately, MMHg only constituted a minor fraction of the Hg speciation in the water samples with a concentration of 0.64 ng l⁻¹. Since Wuli Estuary was heavily contaminated with Hg, elevated total-Hg concentrations in surface water in the Jinzhou Bay were also observed, ranging from 39 to 430 ng l⁻¹. The reported concentrations of total-Hg were two orders of magnitude higher compared to the background concentrations of 0.1–5.0 ng l⁻¹, confirming that this terrestrial area has been seriously contaminated with Hg.

Hg concentrations in seawater of the Xiamen Sea impacted by a coal-fired power plant were recently investigated (Liang et al., 2010). The concentration of total-Hg in surface water samples ranged from 1.5 to 92.9 ng l⁻¹ with a mean value of 11.9 ng l⁻¹, and dissolved Hg from 0.70 to 4.6 ng l⁻¹ with a mean value of 1.5 ng l⁻¹. High concentrations of total-Hg were observed at sites adjacent to the discharge outlet of the plant, which was dominated by particulate-bound species comprising up to 98% of total-Hg. Fu et al. (2010b) revealed that total-Hg concentrations in surface water samples from the South China Sea were in the range 0.80–2.3 ng l⁻¹ with a mean value of 1.2 ng l⁻¹, and MMHg between 0.05 and 0.22 ng l⁻¹, considering that the atmospheric deposition and riverine input were the two primary sources of coastal water Hg, whereas the in situ production by various biotic and abiotic processes may be important sources for MMHg.

3.2. Mercury in sediment

Generally, Hg concentrations in uncontaminated sediments are at similar levels to those observed in background soils, containing less than 1.0 mg kg⁻¹ total-Hg. Su et al. (2004) investigated six lakes in Wuhan City in the central parts of China, and found that total-Hg concentrations in surface sediments (~10 cm) ranged from 0.051 to 0.20 mg kg⁻¹. Similar results were observed in sediments of lakes in the Yangtze Delta and Shanghai City (ranged 0.099–0.13 mg kg⁻¹) (Zhang et al., 2009a). However, sediments impacted by point sources exhibit elevated Hg levels. Sediments of the Baihua Reservoir contaminated by the GOCP plant exhibited elevated Hg concentrations ranging widely from 0.26 to 38.9 mg kg⁻¹ for total-Hg (Yan et al., 2008).

Comparable levels of total-Hg concentrations to uncontaminated reservoirs in sediments from Chinese rivers were reported. In the Huangpu River, for instance, total-Hg concentrations in sediments ranged from 0.071 to 0.39 mg kg⁻¹ (Ding et al., 2005b), compared with

Table 5

Total-Hg concentration in fish from China (mg kg⁻¹, wet weight).

Locations	Range	Mean	N	References
Aquatic around Hong Kong and adjacent mainland waters	0.022–0.14	0.075	83	Liang et al., 2011
The Pearl River Delta	0.0074–0.077		122	Shao et al., 2011
The Pearl River Delta and Hong Kong	0.004–0.067		104	Zhou and Wong, 2000
The Pearl River Delta	0.070–0.34		57	Cheung et al., 2008
Guizhou reservoirs	0.002–0.445	0.066	235	Yan et al., 2010
The Wujiang River	0.005–0.18	0.063	228	Li et al., 2009a
The Three Gorges Reservoir	0.038–0.249		74	Zhang et al., 2007
Lakes from Tibetan Plateau	0.010–0.477	0.145	62	Yang et al., 2011
The Songhua River	0.007–0.082		328	Zhu et al., 2011
Ponds at Wanshan Hg mining	0.061–0.68	0.29	12	Qiu et al., 2009
The Lengshuijiang River impacted by antimony mining area in Hunan	0.0027–0.243	0.035	73	Fu et al., 2010c

0.014 to 0.38 mg kg⁻¹ in the Zaohe River (Jin et al., 2008), and 0.055 to 0.174 mg kg⁻¹ in the Haihe River, respectively (Shi et al., 2005).

Sediments in Chinese coastal areas and estuaries exhibited levels of total-Hg concentrations comparable with those of uncontaminated reservoirs. Total-Hg concentrations in sediments from East China Sea and South China Sea ranged from 0.0041 to 0.20 mg kg⁻¹, and from 0.020 to 0.13 mg kg⁻¹, respectively (Kannan and Falandysz, 1998; Liang et al., 2010; Fang and Chen, 2010). The corresponding concentration intervals for sediments collected from the Pearl River Estuary and the Yellow River Estuary were 0.0015–0.20 mg kg⁻¹, and 0.012–0.12 mg kg⁻¹, respectively (Liu et al., 2009b; Shi et al., 2010). However, high levels of total-Hg in sediments from the contaminated Jinzhou Bay ranged from 0.8 to 25 mg kg⁻¹ with an average of 6.8 mg kg⁻¹, and total-Hg concentrations in sub-samples of the Wuli Estuary were even higher (Wang et al., 2009).

At contaminated sites, high concentrations of MMHg were also detected in sediments, such as 0.5–27.5 µg kg⁻¹ in the Baihua Reservoir (Yan et al., 2008), 8.0–26 µg kg⁻¹ in the Wuli Estuary, and 0.37–5.2 µg kg⁻¹ in the Jinzhou Bay (Wang et al., 2009), respectively. Compared to the data from the contaminated sites, MMHg in sediments of the South China Sea was relatively low at concentrations of 0.010–0.26 µg kg⁻¹ (Kannan and Falandysz, 1998; Liang et al., 2010). Concerning both contaminated and uncontaminated sediments, the proportion of MMHg to total-Hg in samples was typically less than 1%.

3.3. Mercury in fish

Human exposure to Hg occurs mainly from the ingestion of contaminated fish, for which a maximum permissible concentration of 0.5 mg Hg kg⁻¹ has been recommended by WHO (1990). To date, however, very few of the reported data for Hg in Chinese fish exceed the limit of WHO (1990). The concentrations of total-Hg in fish from the Three Gorges Reservoir and the Wujiang River, for example, were generally under the maximum permissible value of 0.50 mg kg⁻¹ (Zhang et al., 2007; Li et al., 2009b). Yan et al. (2010) reported total-Hg concentrations in 12 fish species (n=235) from six reservoirs in Guizhou province, varying in a range from 0.002 to 0.445 mg kg⁻¹, with a mean value of 0.066±0.078 mg kg⁻¹. Hence, none of the fish samples analysed exceeded the WHO limit. Recently, Yang et al. (2011) reported a mean value of 0.145 mg kg⁻¹ (n=62) with a range of 0.010–0.477 mg kg⁻¹ total-Hg for fish collected in lakes of the Tibetan Plateau. Therefore, contrary to fish from North

American and European reservoirs and lakes, fish from Chinese reservoirs and fresh waters seems to have a relatively low Hg content (Table 5).

Concerning the elevated Hg levels observed in fish from North America and northern Europe, Lucotte et al. (1999) suggested that the decomposition of flooded organic matter (OM), which generates favourable conditions for bacterial methylation of inorganic Hg to MMHg, could be the primary driving process underlying high MMHg in fish. It is evident that the conditions in newly constructed Chinese reservoirs with comparatively different type and low availability of organic matters are not in favour of net in-situ methylation (Meng et al., 2010b; Yao et al., 2011; Larssen, 2010). Another important factor is that the most common fish species in Chinese reservoirs are herbivorous species, which grow fast and have a limited time to bioaccumulate MMHg before being consumed.

4. Mercury in the atmospheric environment

Once being released as elemental vapour into atmosphere, Hg will have a residence time of 0.5–2 years corresponding to transportation occurring on a global scale (Munthe et al., 2003). Nevertheless, the physical and chemical properties and transformation processes will determine atmospheric Hg fate and transport (Lindberg and Stratton, 1998). As the biggest consumer of Hg in world, China is being confronted with the issue of huge Hg emissions. For better understanding of the local and/or regional cycling of atmospheric Hg in China, the characterisation of its transportation and deposition is urgently required. A review of Hg emissions and distributions in atmosphere in China is given by Fu et al. (2011) in this issue.

5. Summary

This review highlights the current understanding of Hg in aquatic environment and several hot spot areas in China. China is currently the biggest Hg emitter in the world, and wide ranges and significant variations of Hg concentrations in environmental compartments can be found in urban areas. In remote areas in China, however, the global background levels of Hg can also be observed. Though the results from numerous studies are summarised in this paper as an effort to understand the biogeochemistry of Hg in environments, temporal and spatial distribution of speciated Hg as well as the process of methylation in most ecosystems of China are still not well characterised. In the future, more field and long-term monitoring projects are needed to verify the physical, chemical, and biological processes governing the release of Hg into ecosystems of China.

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