

# MERCURY POLLUTION in China

# An overview of the past and current sources of the toxic metal.

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ercury (Hg) is a toxic element. In the 1960s, residents of Minamata, Japan, developed severe neuropathies from the consumption of methylmercury (MeHg)-contaminated seafood. It is now called Minamata disease, and 2217 cases had been officially recognized in the city by 1989 (1). Hg pollution episodes have also occurred in other countries, such as Iraq, Brazil, Indonesia, the U.S., and China. The U.S. EPA estimates that every year in the U.S. 630,000 infants are born with unsafe levels of Hg in their blood (2). As a result, Hg is now considered a key global pollutant. Because of its persistence, bioaccumulation, and toxicity (PBT) in the environment, Hg is listed as a priority pollutant by many international agencies.

Although some natural emissions of Hg come from volcanoes, forest fires, and soil and water through evaporation, anthropogenic emissions are the main source. A recent global assessment from the UN Environment Programme has documented that the deposition rate of Hg has grown by 1.5-3× during the past century because of increased anthropogenic emissions from industry and agriculture and from medical and domestic sources (3).

China has a long history of mining and using Hg. Two thousand years ago, cinnabar (HgS) was taken by the Chinese because it was believed to prolong life. Cinnabar is still often used today in China as a red pigment, drug, and preservative. With the rapid development of industry and agriculture during the past two decades, the amount of Hg consumed in China has increased significantly. Total Hg (THg) consumption in China was >900 t in 2000, ~50% of worldwide production.

Meanwhile, China is the largest producer and consumer of coal in the world. As of 2003, China consumed nearly 1531 Mt of coal, ~28% of the world's total consumption. China's annual coal consumption is also expected to double to 3037 Mt by 2020 (4). Therefore, China plays an important role in global anthropogenic Hg emissions. The large amount of Hg emitted from China may become a threat to the global environment (5, 6).

The rising anthropogenic emissions have led to serious Hg pollution in China. The aim of this feature is to describe the Hg emissions and pollution in China. In addition, issues related to the current management policy and future research perspectives are discussed.

# Hg emissions and pollution

Asia, especially China, has been regarded as the world's largest atmospheric Hg emission source. Pacyna and Pacyna estimated that in 1995, Asian countries contributed 56% of the worldwide THg emissions, compared with ~30% in 1990 (7, 8). The



Mercury mining residues in Wanshan, Guizhou Province, China.

increase in emissions in Asia was clearly related to the growth of coal combustion in China.

Streets et al. recently compiled an inventory of anthropogenic Hg emissions in China and estimated the country's total emissions at 536 ± 236 t in 1999 (9). Approximately 45% of the Hg comes from nonferrous metal smelting, 38% from coal combustion, and 17% from miscellaneous activities. However, to date, very few studies have been conducted to determine Hg emission factors from different sources in China. As a result, emission factors are typically adopted from studies conducted in Europe and North America with roughly similar sources. Because the processes and pollution-control techniques used in China may differ dramatically from those used in developed countries, these adopted Hg emission factors could differ significantly from the actual field conditions in China. Thus, a large uncertainty could exist in China's anthropogenic Hg emissions inventories.

Mining (gold [Au] and Hg), chemical industry, and coal combustion are the more important anthropogenic sources. The Hg emissions and pollution from these activities are briefly presented next.

# Hg and Au mining

China is rich in Hg mineral resources, ranking third in the world. Most Hg ores are distributed in southwestern and central China, mainly in Guizhou, Shanxi, Henan, and Sichuan Provinces. About 70% of the Hg is found in Guizhou Province, where the metal has been mined for >600 yr. THg emissions to air from mining activities in Guizhou Province alone were ~11 t in 1983 but had declined to ~5 t by 1994 (*10*).

Wanshan, located in eastern Guizhou Province, is known as the "capital of mercury" because it has the largest deposits of Hg in China. The THg produced in Wanshan from the 1950s to the 1990s was ~20,000 t. The city's long history of Hg mining and smelting has resulted in significant pollution of the local ecosystem. Concentrations of Hg in soil range from 24.3 to 348 mg/kg (11), which is 16-232× the maximum Hg concentration allowed for soil in China (1.5 mg/kg, National Standard GB15618-1995). Hg concentrations in river water and plants in the area are also elevated, ranging from 3.2 to 680 ng/L (11-13) and 0.47 to 331 mg/kg, respectively (11). Horvat et al. found that the concentrations of THg and MeHg in rice grains from Wanshan can reach up to 0.57 and 0.14 mg/kg, respectively (12).

Although Hg mining ceased in 2001, >100 Mt of calcines and other waste rocks has been produced as a result of mining in Wanshan. The photograph at left shows mining residues in Wanshan. The Hg concentrations of these residues are relatively low (15–35 mg/kg [14]) compared with those from the mines in Idrija, Slovenia (15); the Humboldt River, Nev., U.S. (16); and the Sulphur Bank mine, Calif., U.S. (17). However, the large amount of mining residues in Wanshan will be a potential source of the toxic metal and a threat to the health of local residents for years.

Au mining that includes amalgamation is considered another important anthropogenic source for Hg emissions. The consumption and emissions of Hg in Au-mining activities have been reviewed by Veiga et al. (18) and Lacerda (19), respectively. In China, Hg is well recovered in large-scale mining processes that use amalgamation, with an emission factor of ~0.79-g Hg/g Au production. However, Hg is poorly recovered in the small-scale (artisanal) mining process, and the emission factor is ~15-g Hg/g Au production (9). In 1999, only ~20 t of Au was produced from large-scale mining processes, and ~16 t of Hg emissions resulted (9). Although artisanal Au-mining activities have been officially prohibited since 1996, a few illegal workshops still operate in remote areas. Because of the high emission factor for artisanal Au mining, Hg emissions from this process are still large; they were ~28.5 t in 1999 (9).

Hg contamination from artisanal Au-mining activities has been studied in Dexing County in Jiangxi Province and Xiaoqinling Region in Shaanxi Province (20-22). According to these studies, ~200 small-scale Au mines used Hg amalgamation in Dexing County during 1990–1995. Many villagers transported gold ore from mines to their houses, which they used as workrooms to extract the metal. As a result, airborne Hg concentrations in the kitchens, bedrooms, and sitting rooms of 4 homes were  $2.4-14.1 \,\mu\text{g/m}^3$ ,  $8-47\times$  the maximum Hg concentration allowed for living environments in China (0.3 µg/m<sup>3</sup>, National Standard TJ 36-1979). In addition, Hg concentrations in the urine of 152 adult villagers in Dexing County ranged from 3.6 to 418 µg/L (average: 65.3 µg/L). About 93.4% of the samples exceeded the normal concentration of Hg in human urine (<20 µg/L) (20).

Many families in Xiaoqinling Region in western China have been involved in small-scale Au-mining activities for a long time. Dai et al. found that the dissolved Hg concentrations in water in Xiaoqinling Region ranged from 0.11 to 3.1 µg/L (average: 0.74 µg/L), and the particulate Hg (Hg<sup>P</sup>) concentrations were much higher (0.1–259 µg/L) (21). THg concentrations in vegetable and wheat samples varied from 41.7 to 636 µg/kg; they all significantly exceeded the Chinese guidance limits for vegetables (10 µg/kg) and foodstuffs other than fish (20 µg/kg) (22). These results indicate that Hg pollution from small-scale Au-mining activities may still be serious and that further studies are needed.

# **Chemical industry**

Because of its special physical and chemical characteristics, Hg has been widely used in numerous industrial processes and products, such as chlor–alkali production, batteries, fluorescent lamps, thermometers, and electronic switches. In China, the amount of Hg consumed has increased significantly during the past two decades. Hg has been well recovered in most chemical plants in recent years, and the emissions of Hg from the chemical industry are relatively low (23); however, serious Hg pollution caused by wastes discharged from chemical plants occurred in some regions, such as the Songhua and Jiyun Rivers.

The Songhua River in northeastern China is well known for an industrial spill that polluted it with nitrobenzene in November 2005. In fact, serious Hg pollution also occurred in the Songhua River during the 1970s and 1980s, mainly caused by wastewater discharged from a plant of the Jinlin Chemical Co. The plant was the largest producer of acetaldehyde in China at that time and used  $HgSO_4$  as a catalyst to produce the chemical. The process is basically the same as that used by the Chisso Co. in Minamata, which eventually led to the infamous outbreak of Hg poisoning.

Wastewater from the Jinlin chemical plant contained high concentrations of inorganic Hg and MeHg and was directly discharged to the Songhua River. At peak operation between 1973 and 1976, the average input rates of THg and MeHg from the wastewater were 33 and 1.5 kg/d, respectively. The average concentration of THg in fish muscles was 0.74 mg/kg, and the highest value was 3.24 mg/kg (24). The average Hg concentration in the hair of local fishermen reached 13.5 mg/kg, and several showed symptoms of Minamata disease (25). After 1977, the discharge of Hg was controlled, and the input rates of THg and MeHg decreased to 3 and 0.2 kg/d, respectively. The THg concentration in fish muscles dropped to 0.44–0.54 mg/kg.

In 1982, the old technique for producing acetaldehyde was abandoned, and Hg was no longer used in this plant. Hg concentrations in fish and fishermen's hair have since decreased substantially. For instance, levels of Hg in hair dropped to 0.2–2.3 mg/kg in 2002, a reduction of 89% from the early 1990s (*26*). In 2003, the THg concentrations in fish muscles were 0.05–0.27 mg/kg (*27*). However, ~150 t of Hg had been discharged into the Songhua River from 1958 to 1982. It had been incorporated in the sediments and represents a long-term source of Hg to the local ecological system. The releases of Hg over time from the river sediments will continue to be an environmental and health concern to the local residents.

Chlor-alkali production is the largest intentional



The Beijing Shi Jing-shan power plant is one of China's many sources of airborne mercury.

use of Hg in the world. The global Hg consumption for chlor–alkali production was ~1344 t in 1996, 40% of the THg consumed that year (3). Hg used in chlor– alkali production can be released into the environment via atmospheric emission, water discharges, and sodium hydroxide and hydrogen products. The THg released from the chlor–alkali industry in China has been ~1400–2700 t since the 1950s (28). An estimated 17.5–20% of the Hg was released into water, 49.1– 53.3% was discharged to the brine system outside the plants, 10–14% was emitted directly to the atmosphere, and 5.3–6.7% entered waste materials (28).

The Jiyun River is a typical example of Hg pollution caused by chlor-alkali production. It is located in Hebei Province and is an important river in eastern China. It has been seriously contaminated with Hg since the 1960s when a chlor-alkali plant was built along its middle reach. Zhang et al. investigated Hg concentrations in the water and sediment of the Jivun River in 1977 (29). They showed that the Hg levels were low upstream of the plant but extremely high downstream. The highest Hg concentrations were found in water and sediment near the plant, at 24 µg/L and 1470 mg/kg, respectively. Huang et al. reported that the Hg concentrations in planktons, benthos, herbivorous fish, omnivorous fish, carnivorous fish, and waterfowl in the Jiyun River estuary were 0.34, 0.48, 0.90, 1.28, 1.70, and 3.29 mg/kg, respectively (30). After 1977, a control project was implemented, and Hg concentrations in wastewater decreased tremendously (29).

# **Coal combustion**

Coal combustion is the most important source of global atmospheric Hg. About 75% of the THg emissions are attributed to the combustion of fossil fuels, particularly coal combustion in China, India, and South and North Korea (7, 8). Hg in coal is discharged to the atmosphere, ash, and cinder. Atmospheric Hg can be transported over a long distance and deposited far from its origin. In addition, residents burning coal in rural homes can be exposed to elevated levels of Hg when they use unventilated stoves and when they don't wash the coal.

Wang et al. reported that the average Hg concentration of coal in China is 0.22 mg/kg; thus, they estimated that THg emission from coal combustion in 1995 was 303 t. About 210 t was emitted to the atmosphere, which is >5% of the global Hg emissions (*31*). Wang et al. also estimated that China had emitted > 2400 t of Hg into the atmosphere from 1978 to 1995 through coal combustion, with Hg emissions increasing at an average rate of 4.8%/yr (*31*).

However, Huang et al. thought that the average Hg concentration of coal in China should be 0.15 mg/kg, on the basis of analytical results from 1466 coal samples from around the country (32). According to these data, Hg emission from coal combustion in China is much lower than that estimated by Wang et al. (31).

Recently, Streets et al. estimated that 202 t of Hg was emitted from coal combustion in 1999, including 33.1 t of Hg<sup>0</sup>, 124 t of Hg<sup>2+</sup>, and 45.4 t of Hg<sup>P</sup>. Of the Hg emitted from coal combustion in China,

# FIGURE 1

# Mercury emissions from coal combustion in 1999 by province

Data from Ref. 9.



51% comes from industrial use, 33.6% from power plants, 9.8% from residential use, and 5.6% from other uses (9).

Hg emissions from coal combustion in 1999 are shown by province in Figure 1. Hg emissions in central-eastern and southwestern China were obviously most significant.

Coal combustion has resulted in elevated Hg concentrations in the air of some cities. Changchun is a typical metropolitan city in northeastern China, with a population of 2.92 million. Winters in Changchun are long and cold. Coal is the main energy source in the city, and ~7.1 Mt was consumed in 2001 (33). During the 1999–2000 heating period, the average concentration of total gaseous Hg (TGM) in Changchun was 25.4 ng/m<sup>3</sup>. During the nonheating period, the average concentration of TGM in Changchun dropped to 13.5 ng/m<sup>3</sup> (33). The TGM concentrations in the heating season were significantly higher than those in the nonheating season. The reported concentrations of TGM in Beijing, Guiyang, and Chongqing were 6-10 (34), 7.4 (35), and 34.4 ng/m<sup>3</sup> (36), respectively. All these TGM concentrations are much higher than the global background concentration of  $1.5-2.0 \text{ ng/m}^3$  (37).

Atmospheric Hg can accumulate in surface soil and water through wet and dry deposition. More than 90% of it enters the terrestrial ecosystem (*37*, *38*), and soil is the largest Hg recipient (*39*). A survey conducted by the China National Environmental Monitoring Center found that the national average concentration of Hg in soil was 0.038 mg/kg (*40*). However, in Beijing, Chongqing, Guiyang, Guilin, Guangzhou, Nanjing, and Taiyuan, Hg concentrations in urban soils are quite high; the highest concentration is 11× higher than the average concentration in China (*41*). This pattern is closely related to the coal combustion in these cities.

# **Contamination in food**

For the general population, dietary intake is the

dominant exposure pathway to Hg. Seafood is considered the main source of MeHg in the human diet. Studies have shown that 75–95% of Hg in most adult fish exists as MeHg. In 1997, EPA set an MeHg guideline for the diet at 0.1 µg of Hg per kg of body weight per day (42). The Food and Agriculture Organization of the UN and the World Health Organization recently lowered the provisional tolerable weekly intake for MeHg from 3.3 to 1.6 µg/kg of body weight to provide sufficient protection for human health (43). The tolerance limit for Hg in seafood in China is 0.3 mg/kg for THg and 0.2 mg/kg for MeHg (National Standard GB2762-1994).

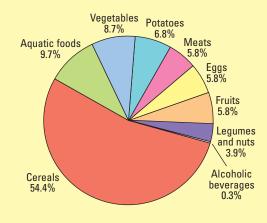
However, THg and MeHg concentrations in seafood in China are largely unknown. Recently, Liang et al. investigated the THg and MeHg contamination in mollusk samples collected from eight cities along the Bohai Sea: Dalian, Yingkou, Huludao, Qinhuangdao, Tanggu, Yangkou, Penglai, and Weihai (44). The results showed that THg concentrations in the samples were 0.01–0.45 mg/kg. Although some of the samples tested higher than the tolerance limit for THg in seafood in China, MeHg concentrations were relatively low at 0.01–0.17 mg/kg. MeHg made up 19–89% of the THg, which is in accordance with that in mollusk from the French coast (45) and the Krka Estuary (Croatia) (46).

Chen and Gao reported that the dietary intake of THg for Chinese residents was 10.3 µg/d in 1990 (47). This value was higher than those in Finland, Germany, Sweden, The Netherlands, the U.K., and the U.S. but lower than those in Brazil, Croatia, Japan, Poland, and Spain (3). The percentages of Hg from different food groups in Chinese dietary THg intake are shown in Figure 2. Cereals and cereal products are the largest source of dietary Hg intake in China (54%). Aquatic foods contribute only 9.7%, which is less than for many other countries. However, it should be noted that seafood consumption is still a major intake source in some coastal areas of China (48, 49).

# FIGURE 2

# Percentages of mercury in the Chinese diet from different food groups

Total mercury intake in 1990. Data from Ref. 47.



# **Current policy and future perspectives**

Reducing Hg pollution has been a high priority in China's environmental management and improvement program. During the past few years, China's State Environmental Protection Administration has also strengthened its management of the production, use, import, export, and disposal of Hg. Hg mining has been restricted in many areas, such as Wanshan. In 1996, artisanal Au-mining activities were officially banned.

Since the late 1970s, most chlor–alkali production processes have been improved, closed, or converted to Hg-free technologies. The Hg loss factor from the chlor–alkali industry was reduced considerably from 500–1400 g/t NaOH production before 1977 to 160–180 g/t NaOH in 1997. The number of Hg cells in chlor–alkali plants has also been reduced. In 1975, ~10% of the plants used Hg cells, whereas only 2% used them in 1995 (*28*). Hg emission from coal combustion has also been slightly reduced in recent years, because the government imposed regulations in 1998 on the sulfur content of coal used in urban environments and clean coal technology continues to improve (*50*).

However, studies on Hg pollution in China are inadequate in four important areas. First, data on the production, consumption, and emission of Hg are still very limited. Monitoring of Hg levels in various environmental media and establishing a national database on the fluxes of Hg in China are necessary. Second, Hg levels in seafood in China are still largely unknown, although a tolerance limit has been established. A more detailed consumption advisory is urgently needed. Third, because coal combustion is the main source of anthropogenic Hg, studies of Hg emissions from coal combustion should be carried out, and strict regulations are needed. Finally, reducing the volume of Hg used is as important as legislation controlling its emissions. Therefore, safer alternatives and cleaner technologies must be developed and effectively implemented in China.

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### Disclaimer

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