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Anthropogenic mercury emissions in China

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Abstract

An inventory of mercury emissions from anthropogenic activities in China is compiled for the year 1999 from official statistical data. We estimate that China's emissions were 536 ($\pm$236) t of total mercury. This value includes open biomass burning, but does not include natural sources or re-emission of previously deposited mercury. Approximately 45% of the Hg comes from non-ferrous metals smelting, 38% from coal combustion, and 17% from miscellaneous activities, of which battery and fluorescent lamp production and cement production are the largest. Emissions are heaviest in Liaoning and Guangdong Provinces, where extensive smelting occurs, and in Guizhou Province, where there is much small-scale combustion of high-Hg coal without emission control devices. Emissions are gridded at 30×30 min spatial resolution. We estimate that 56% of the Hg in China is released as Hg^0, 32% as Hg^{2+}, and 12% as Hg^p. Particulate mercury emissions are high in China due to heavy burning of coal in residential and small industrial settings without PM controls. Emissions of Hg^{2+} from coal-fired power plants are high due to the absence of flue-gas desulfurization units, which tend to dissolve the soluble divalent mercury. Metals smelting operations favor the production of elemental mercury. Much of the Hg is released from small-scale activities in rather remote areas, and therefore the activity levels are quite uncertain. Also, emissions test data for Chinese sources are lacking, causing uncertainties in Hg emission factors and removal efficiencies. Overall, we calculate an uncertainty level of $\pm$44% (95% confidence interval) in the estimate of total emissions. We recommend field testing of coal combustors and smelters in China to improve the accuracy of these estimates.

Keywords: Speciated mercury; Emission inventory; Coal combustion; Metals smelting; China

1. Introduction

Concern about mercury in the environment has grown to the point where action is believed to be warranted to reduce the risks to humans and wildlife (US EPA, 1997; UNEP, 2002). Environmental mercury levels have increased considerably in recent years; and even regions with no significant emissions, such as the Arctic, are affected by the transcontinental transport of mercury. Modeling studies have confirmed the ability of elemental

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mercury to be transported over long distances (Seigneur et al., 2001; Travnikov and Ryaboshapko, 2002; Banic et al., 2003; Dastoor and Larocque, 2004). This has generated concern in the United States that the quantities of imported atmospheric mercury may be substantial and may interfere with the ability of domestic sources to comply with future emission limitations (Steding and Flegal, 2002; Seigneur et al., 2004). Seigneur et al. (2004) estimate that anthropogenic emissions of mercury in Asia contributed 21% to total mercury deposition in the contiguous United States in 1998.

Global emissions of anthropogenic mercury to the atmosphere have been estimated to be 1900 t (all emissions in metric tons, or t) in 1995 (Pacyna and Pacyna, 2002), of which 77% was from stationary fuel combustion, with the remainder divided among non-ferrous metals production, cement production, and waste disposal. Pacyna and Pacyna (2002) estimated that China's emissions from coal combustion contributed more than 25% to total global emissions. This inventory built heavily on experience in Europe and North America (Pirrone et al., 2001; Pacyna et al., 2001), and the authors acknowledge higher uncertainty for other continents. The inventory was gridded at $1^\circ \times 1^\circ$ resolution (Pacyna et al., 2003). Of total global anthropogenic emissions, Pacyna and Pacyna (2002) estimated that 53% was emitted as elemental mercury (Hg^0), 37% as gaseous divalent mercury (Hg^{2+}), and 10% as particulate mercury (Hg^p). More recent assessments of the anthropogenic mercury contribution raised the global estimate to 2000–2200 t yr^{-1} (Seigneur et al., 2004). In addition to anthropogenic emissions, natural emissions from land and ocean are thought to be on the order of 2000 t yr^{-1}, and the re-emission of anthropogenic mercury previously deposited is thought to be about 2000 t yr^{-1}, yielding a total input to the environment of about 6000–6500 t yr^{-1} (Bergan et al., 1999; Mason and Sheu, 2002; Seigneur et al., 2004).

Mercury contamination is a serious problem in China, particularly in the Southwest. Feng (2005) has summarized a number of specific instances associated with industrial releases of mercury in past years: contamination of the Songhua River in Jilin Province by an acetic acid plant, contamination of the Jiyun River in Tianjin by a chlor-alkali plant, contamination of the Dongmenqiao River in Guizhou Province by an acetic acid plant, and contamination of the Yellow River by a chlor-alkali plant in Gansu Province. Fortunately, these large-scale problems were detected and emissions were eliminated by process changes or plant closures, which is not to say that problems do not persist in these areas because of the accumulated inputs of mercury to the local ecosystems. And though few large point-source emissions of atmospheric mercury remain in China, dispersed emissions continue at high levels because of the large quantities of coal burned and the extent of other mercury-releasing activities.

High concentrations of mercury in the air of China's cities have been reported in several studies. Average concentrations of total gaseous mercury in Guiyang, Guizhou Province, have been measured in the range of 5–15 ng m^{-3} (Feng et al., 2002, 2003, 2004a, b), attributed to uncontrolled coal-burning in the residential and industrial sectors. In Beijing, Liu et al. (2002) measured total gaseous mercury concentrations in the range of 6–10 ng m^{-3} during winter. Fang et al. (2001) measured average particulate mercury concentrations of about 0.5 ng m^{-3} in northeastern Changchun City, Jilin Province, rising to as high as 2 ng m^{-3} during the heating season. Xiu et al. (2005) measured somewhat lower levels of Hg in TSP in Shanghai, in the range of 0.2–0.5 ng m^{-3}.

There is only one published estimate of mercury emissions by Chinese researchers, which has been reported in three papers (Wang et al., 1999, 2000; Zhang et al., 2002). The estimate is 214 t of mercury released from coal combustion in 1995. Pacyna and Pacyna (2002) did not report calculated emissions from China per se but estimated that Asian emissions in 1995 were 860 t from stationary fuel combustion (primarily coal) and 1074 t in total and suggested that China's emissions were approximately 500 t. This previous body of work is inadequate because it is either incomplete or lacking in local knowledge of mercury sources.

In this paper, we present a comprehensive inventory of mercury emissions from anthropogenic sources (no natural sources or re-emission) in China for the year 1999. Following the precedent of our Asian TRACE-P emission inventory (Streets et al., 2003a, b), we include emissions from open biomass burning, arguing that the majority of such burning is directly or indirectly attributable to human activities. We develop a detailed assessment of emissions from coal combustion with a new technology-based treatment for each province, supplemented with estimates of emissions from all other significant man-made sources. Mercury

emissions are speciated using technology-specific factors.

2. Methodology

A model has been developed to calculate anthropogenic Hg emissions in China. Emissions are calculated using fuel consumption data and detailed Hg emission factors. The basic concept of the Hg emission calculation is described by the equation

$$E = \sum_i \sum_j [\mathrm{ef}_{i,j} A_{i,j} F_{\mathrm{REL}_j} (1 - F_{\mathrm{REM}_j})], \quad (1)$$

where E is the Hg emission; $\mathrm{ef}_{i,j}$ is the Hg content of coal as burned or emission factor for other fuels or non-combustion processes; $A_{i,j}$ is the amount of fuel consumption or production yield of non-combustion processes; F_{REL_j} is the fraction of Hg released to the atmosphere; F_{REM_j} is the fraction of Hg removed by emission control devices; j is the combustor type with/without emission control devices; and i is the province.

Fuel consumption data by sector and fuel type are provincial-level estimates compiled from the China Energy Statistical Yearbook for 1999 (NBS, 2001). Material yield data (e.g., cement production) related to non-combustion processes by province are from the China Statistical Yearbook (NBS, 2000) and professional yearbooks such as the China Nonferrous Metals Industry Statistical Yearbook (ECCNMI, 2000). Thirty-seven source categories are included in this inventory, covering all significant combustion and non-combustion activities in China. Many of the fuel/sector combinations are sub-divided into different types of combustors and/or different types of emission control devices (Streets et al., 2001).

2.1. Mercury content of coal and coal products

Several studies (USGS, 2004; ITPE, 2003; Huang and Yang, 2002; Wang et al., 2000; Zhang et al., 1999; Ni et al., 1998) report the Hg content of raw coal as mined in China, among which significant differences are shown in Table 1. These differences can be attributed to selection of coal mine, location of sampling, test methodology, etc. Considerable heterogeneity of mercury content in different coal seams is well known. We merged the USGS data and the Chinese literature data for our study, as shown in Table 1. As a result, the averaged Hg content by production in China is 0.19 mg kg^{-1} in 1999. Work is continuing by the US Geological Survey to try to reconcile the disparity of measurements within provinces.

The Hg content of cleaned coal as produced is calculated as follows:

$$\mathrm{ef}_{\mathrm{cc},i} = \frac{\mathrm{ef}_{\mathrm{rc},i} C_{\mathrm{rc},i} (1 - F_{\mathrm{REM}})}{P_{\mathrm{cc},i}}, \quad (2)$$

where $\mathrm{ef}_{\mathrm{cc},i}$ is the Hg content of the cleaned coal; $\mathrm{ef}_{\mathrm{rc},i}$ is the Hg content of the raw coal; $C_{\mathrm{rc},i}$ is the amount of raw coal consumption; $P_{\mathrm{cc},i}$ is the amount of clean coal production; F_{REM} is the fraction of Hg removed by the coal cleaning process; and i is the province. The Hg removal efficiency of coal cleaning can vary significantly from ~10% to ~80% (Wang and Peng, 2003; Feng et al., 2001; Luttrell et al., 2000; US EPA, 1997; Akers, 1996). Most estimates are between 25% and 60%. Based on the characteristics of coal cleaning methods in China, we assume an average removal efficiency of 30% independent of mercury content.

In China, coal briquettes and coke are produced from both raw coal and cleaned coal. Calculation of the Hg content of briquettes and coke, as produced, is based on Eq (3):

$$\mathrm{ef}_{\mathrm{b/c},i} = \frac{(\mathrm{ef}_{\mathrm{rc},i} C_{\mathrm{rc},i} + \mathrm{ef}_{\mathrm{cc},i} C_{\mathrm{cc},i})(1 - F_{\mathrm{REM}})}{P_{\mathrm{b/c},i}}, \quad (3)$$

where $\mathrm{ef}_{\mathrm{b/c},i}$ is the Hg content of the briquettes or coke; $C_{\mathrm{cc},i}$ is the amount of cleaned coal consumption; $P_{\mathrm{b/c},i}$ is the amount of briquettes or coke produced; and F_{REM} is the fraction of Hg removed by the briquette production or coking process.

Most of the mercury is released into the atmosphere or to other co-products during the coking process. Hong et al. (2002) indicated that 10.7% of Hg remained in coke in their test samples, and a similar result of 14.5% of Hg remaining in coke and other co-products was found by Wang et al. (2000). We assume that 10% of the Hg remains in the coke after the coking process. Because no evidence shows that there is Hg removal during the briquette production process, we assume that 100% of the mercury in the raw coal or cleaned coal is transferred to the briquettes.

2.2. Mercury content of coal as burned by province

In order to obtain reliable estimates of the magnitude and spatial distribution of Hg emissions, it is essential to know the Hg content of the coal as burned, not just as mined. Therefore, it is necessary

Table 1
Hg content of raw coal as mined in China by province ($mg\,kg^{-1}$)

Province[a]	This study[b]	USGS, 2004	Wang et al., 2000	Huang and Yang, 2002	ITPE, 2003	Zhang et al., 1999	Ni et al., 1998
Anhui	0.26	0.19	0.22	0.26	0.37		
Beijing	0.44	0.54	0.34				
Fujian	0.08	0.07			0.08		
Gansu	0.05	0.05					
Guangdong	0.15	0.05			0.25		
Guangxi	0.30	0.33			0.28		
Guizhou	0.52[c]	0.20		0.52	0.14	0.55	0.50
Hainan	0.15[d]						
Hebei	0.14	0.14	0.13	0.80[e]			
Heilongjiang	0.09	0.06	0.12	0.14[e]			
Henan	0.25	0.21	0.30	0.17	0.32		
Hubei	0.16	0.16					
Hunan	0.10	0.14		0.07			
Jiangsu	0.16	0.34		0.09	0.04		
Jiangxi	0.22	0.27	0.16				
Jilin	0.20	0.07	0.33				
Liaoning	0.17	0.18	0.20	0.13			
Nei Mongol	0.22	0.15	0.28	0.02[e]	0.63		
Ningxia	0.20	0.20					
Qinghai	0.04	0.04					
Shaanxi	0.11	0.13	0.16	0.08	0.07		
Shandong	0.18	0.13	0.17	0.21	0.22		
Shanxi	0.16	0.15	0.22	0.20	0.07	0.16	
Sichuan	0.14	0.11	0.18				
Xinjiang	0.02	0.02	0.03				
Yunnan	0.29	0.14		0.34		0.38	
Zhejiang	0.35				0.35		
China	0.19[f]						

[a]Hong Kong, Macao, Shanghai, Tianjin, and Xizang do not produce raw coal and are not included in this table.
[b]The USGS and Chinese literature data are merged in our study.
[c]Because the largest Hg mine deposit in China is in Guizhou province, only the higher Hg contents in coal from Huang and Yang, (2002), Zhang et al. (1999) and Ni et al. (1998) are used.
[d]The value is assumed to be equal to that for Guangdong province due to lack of samples.
[e]Only one sample for each province was tested.
[f]The data are averaged by raw coal production of each province in 1999.

to relate the coal produced (mined) in particular provinces to its consumption in each province. According to statistical data from CCTA (2003), NBS (2001), and other literature (Wang, 2001; Jiang, 2004), a transportation matrix by coal type (e.g., raw coal, cleaned coal) and by sector (e.g., power plants, industrial use) was set up to quantify in-province coal use and inter-province coal flows. Then the Hg content of coal for the consuming provinces is calculated as follows:

$$ef_{c,i,j} = M_{i,j} ef_{s,i,j}, \quad (4)$$

where $ef_{c,i,j}$ is the Hg content of coal in the consuming provinces; $M_{i,j}$ is the transportation matrix; $ef_{s,i,j}$ is the Hg content of coal in the supplying provinces; i is the coal type (raw coal, cleaned coal, coke, briquette); and j is the sector (power, coking, industrial, residential, other uses).

Table 2 presents the estimated Hg content of raw coal as burned in China by province and sector in 1999; the Hg content of raw coal as produced is listed for comparison. We also estimated the Hg content of cleaned coal, coke, and briquettes as burned by province, but these values are not presented here. For the major coal supplying provinces, such as Shanxi, Shaanxi, Nei Mongol, and Guizhou, there is no or very little difference between the Hg content of coal as produced and as

Table 2
Comparison of Hg content of raw coal as produced and as burned in China by province in 1999 (mg kg^{-1})

Province	Coal as produced	Coal as burned			
		Power plants	Coking	Industry	Others
Anhui	0.26	0.23	—	0.24	0.24
Beijing	0.44	0.24	0.16	0.26	0.31
Fujian	0.08	0.11	0.08	0.12	0.08
Gansu	0.05	0.07	0.05	0.06	0.05
Guangdong	0.15	0.18	—	0.19	0.16
Guangxi	0.30	0.28	—	0.32	0.28
Guizhou	0.52	0.52	0.52	0.52	0.52
Hainan	0.15	0.16	—	0.21	0.15
Hebei	0.14	0.15	0.15	0.16	0.15
Heilongjiang	0.09	0.14	0.09	0.09	0.09
Henan	0.25	0.23	0.23	0.24	0.25
Hong Kong	—	0.18	—	—	—
Hubei	0.16	0.18	—	0.19	0.17
Hunan	0.10	0.13	0.10	0.13	0.10
Jiangsu	0.16	0.16	0.16	0.19	0.18
Jiangxi	0.22	0.22	—	0.22	0.22
Jilin	0.20	0.20	—	0.16	0.18
Liaoning	0.17	0.18	—	0.16	0.17
Macao	—	—	—	—	—
Nei Mongol	0.22	0.20	0.21	0.20	0.22
Ningxia	0.20	0.19	—	0.20	0.20
Qinghai	0.04	0.04	—	0.04	0.04
Shaanxi	0.11	0.11	0.12	0.12	0.11
Shandong	0.18	0.18	0.18	0.18	0.18
Shanghai	—	0.18	—	0.24	0.23
Shanxi	0.16	0.16	0.16	0.16	0.16
Sichuan	0.14	0.18	0.14	0.14	0.14
Tianjin	—	0.16	—	0.22	0.23
Xinjiang	0.02	0.02	0.02	0.02	0.02
Xizang	—	—	—	—	—
Yunnan	0.29	0.32	0.29	0.30	0.29
Zhejiang	0.35	0.17	—	0.24	0.27

burned, because all or nearly all of the coal is obtained from within-province supply. However, for provinces such as Beijing, Guangdong, and Zhejiang, where most of the consumed coal originates in other provinces, there is a significant difference between the Hg content of coal as produced and as burned.

2.3. Combustor types and emission controls by sector

Because the Hg release rates and the speciation profiles depend greatly on combustion technology and conditions, it is necessary to develop a detailed specification of the ways in which coal is burned in China. Our model contains 65 individual source types for coal combustion, 22 of which are for coal-fired power plants, 30 for industrial use, 9 for residential use, and 4 for other uses including construction, commerce, transportation, and so on. The partitioning of each combustion technology/control device/fuel type by province and sector is built into our model based on a wide literature review (CCTA, 2003; CRAES, 2003; Wang, 2003a; ECCCEY, 2000; ECCMIY, 2000; MOA, 2000; Yan, 1997; MPI, 1996; SEPA, 1996).

Coal-fired power plants consumed about one-third of the total coal in China in 1999. At present, pulverized-coal (PC) boilers are widely used in power plants in China, representing over 90% of the market share. The remaining share is represented by stokers, which are mainly used in small coal-fired power plants. Since the mid-1980s, electrostatic precipitators (ESP) have been increasing their share by 4–5% annually to replace wet particle scrubbers and cyclones in power plants (Wang, 2003b); now the share of ESPs has reached more than 50% and is as high as 90+% in provinces such as Beijing.

Industry is the biggest coal-consuming sector in China, burning 30% of the raw coal, 15% of cleaned coal, 9% of briquettes, and 90% of coke in 1999. Far different from the power-plant sector, stokers are the dominant boiler types used in the industry sector in China, contributing over 90% of the market share. Fluidized-bed furnaces (FBF) are another important boiler type with share of about 8%. Cyclones and wet scrubbers are the two main types of PM control device used on industrial boilers, contributing nationally averaged 61% and 28% of market share, respectively. However, there are still ~10% of industrial boilers without any PM control in China; and for some poorer and more remote provinces such as Guizhou and Yunnan that share reaches as high as ~30%.

Residential use is also an important coal-consuming sector in China, representing 7% of raw coal, 5% of cleaned coal, and 90% of briquettes in 1999. Traditional cookstoves and improved cookstoves are the major combustion types for residential cooking and heating, both of which are without any PM control device. In the big cities, however, many residents obtain heat from centralized heating systems that use mid- or large-sized boilers. This part of the coal consumption for residential heating use assumes the use of stoker boilers with cyclone controls. For farming, construction, transportation, and commerce, the coal consumption is combined and assumes the use of small stokers without any PM control.

A fraction of the mercury contained in the fuel is not emitted to the air but is retained in the bottom ash and disposed of as solid waste. The share of Hg remaining in the bottom ash is different for different boiler types. Many studies (Huang et al., 2003; Meij, 1991; US EPA, 2002; Zhu et al., 2002) indicate only 1–2% of Hg remains in the bottom ash for PC boilers in power plants; however, the ratio may increase to about 7–9% for industrial PC or FBF boilers and 17–18% for industrial stokers (Wang et al., 2000; Wang and Ma, 1997). In our study, the shares of Hg remaining in the bottom ash for different boiler types are listed in Table 3.

Control technologies used to reduce criteria air pollutant emissions (e.g., particulate matter) from combustion boilers also remove some of the Hg from the flue gas; however, the removal efficiencies vary widely. Table 4 presents assumed Hg removal efficiencies of the three predominant types of PM control devices installed in boilers in China: ESP has a moderate Hg removal efficiency of ~30%; wet PM scrubbers show very little benefit, with Hg removal efficiency under 10%; and cyclones remove essentially no mercury. However, there is very little information about Hg removal efficiencies on devices other than ESPs on PC power plants. In addition, there are almost no data at all for China—most of the tests to date have been conducted in the US or Europe. Fortunately, Zhejiang University has recently begun a program of testing Hg emissions from Chinese sources.

Table 3
Share of Hg remaining in bottom ash for major boiler types by sector

Sector	Boiler type	Ratio (%)
Residential use	Traditional cookstove	17.0
	Improved cookstove	17.0
	Stoker	17.0
Industrial use	FBF/PC	8.0
	Stoker	17.0
Power plant	PC	1.0
	Stoker	2.0
Farming, construction, etc.	Stoker	17.0

2.4. Hg emission factors for other sources

Besides coal combustion, there are many other combustion sources (other fossil fuels, biomass, municipal waste, etc.) and non-combustion sources (cement production, ferrous and non-ferrous metals smelting processes, caustic soda production, etc.) contributing to Hg emissions in China. Emission factors for these other sources used in the 1999 emission inventory for China are listed in Table 5. Hg emission factors for spontaneous burning in coal mines and for zinc smelting are at provincial level; however, only nationally averaged emission factors are available for most of the other source types.

2.5. Speciation of Hg compounds

Primary emissions are classified according to gaseous elemental mercury (Hg^0), divalent gaseous mercury (Hg^{2+}), and particulate Hg (Hg^p). In the absence of China-specific speciation data, we use measurements by the US EPA (2002), Friedli et al. (2001, 2003a, b) and Pacyna and Pacyna (2002) for coal combustion sources, vegetation burning, and other sources, respectively, as shown in Table 6. The lack of speciation measurements on the particular types of combustors prevalent in China, particularly stokers and cookstoves, hinders the development of accurate profiles of species at national and provincial scales.

3. Emissions by sector

Total Hg emissions by sector associated with fuel consumption (e.g., coal, fuel oil, and biofuels) and material yields (e.g., cement production, zinc production) are summarized in Table 7 and

Table 4
Hg removal efficiencies of various PM control devices (%)

	This study	US EPA, 2002	US EPA, 1997	Wang et al., 2000	Zhu et al., 2002
ESP	30.6	36.0	30.4	25.7	30.3
PM scrubber	6.5	8.7	4.3		
Cyclone	0.1	0.1	0.0		

discussed below. Furthermore, total Hg emissions are classified into Hg^0, Hg^{2+}, and Hg^p, respectively, by each sector, also as shown in Table 7.

3.1. Coal combustion emissions

In 1999, China consumed 1197.3 million tons of raw coal, 163.1 million tons of cleaned coal, 92.3 million tons of coke and 9.7 million tons of briquettes, for power plants, industrial use, residential use and other uses (NBS, 2001). Such a huge amount of coal consumption resulted in 202.4 t of total Hg emissions, with a speciation split as follows: 33.1 t of Hg^0, 123.9 t of Hg^{2+}, and 45.4 t of Hg^p. Coal-fired power plants consumed 487 million tons of raw coal in China in 1999. We estimate that total Hg emissions were 68.0 t in 1999. Hg^{2+} is the majority species from power plants due to the absence of FGD on most power plants, which takes up soluble Hg^{2+}. ESP technology has been increasing its share to replace wet scrubbers and cyclones in power plants since the mid-1980s, and now its share has reached over 50%. Based on current ESP installation capacity, we estimate that about 16 t of Hg were removed in 1999 in the power sector.

In 1999, industrial coal use was also very large (538 million tons of raw coal and 228 million tons of other coal products) and resulted in the emission of 103.2 t of total Hg. Industrial use is the biggest single sector, contributing 50.7% of the total Hg emissions in the coal-combustion category. This is attributed to two major reasons: (1) it is the largest coal-consuming sector in China; and (2) it contains a significantly higher share of uncontrolled or poorly controlled boilers compared to the power-plant sector. Currently, most of the cleaned coal in China is used in the industrial sector, and the majority is for coking. However, the share of cleaned coal to total coal consumption, about 15%, is much lower than in other countries. We estimate that coal cleaning removed about 12 t of Hg in this sector in 1999.

Residential coal use amounted to 123 million tons of coal products and emitted 19.7 t of total Hg in 1999. The residential sector contributed only 10% of total Hg emissions in the coal-combustion category. The majority of emissions are in the form of Hg^p in this sector, with the share reaching 88% of total, because most of the coal in this sector is burned in small, low-temperature domestic stoves, cookers, and heaters without any emission controls. Emission of fine particles is greatly enhanced under these conditions, which favor gas-to-particle conversion of gaseous mercury, thus yielding relatively more emissions of Hg^p. Most briquettes are consumed in households, and the demand for briquettes is increasing; however, no mercury reduction benefit accrues because no evidence supports Hg removal during the briquette production process. Coal consumption for farming, construction, transportation, and commerce is combined together as other uses (71 million tons of coal products used). We estimate that total Hg emissions were 11.5 t in 1999, mostly particulate Hg, similar to the residential sector.

3.2. Non-ferrous metals smelting emissions

Non-ferrous smelting operations are known to be a large source of mercury, especially in less developed parts of the world. For example, artisanal activities to liberate gold often use a mercury amalgamation technique with evaporation of Hg from the amalgam as the final step. This leads to dangerous atmospheric releases of mercury (Gunson and Yue, 2001; Gunson and Viega, 2004; Lin et al., 1997). Zinc, lead, copper, and gold smelting are estimated to emit 242.4 t of total Hg in 1999, with speciation split as follows: Hg^0 193.9 t, Hg^{2+} 36.4 t, and Hg^p 12.1 t.

Zinc production in China is increasing significantly, reaching 1.7 million tons in 1999. We estimate that total Hg emissions in 1999 were 147.6 t. In our study, the Hg emission factor for zinc smelting varied from 13.8 to 156.4 $g\,t^{-1}$ Zn, with a nationally averaged emission factor of 86.6 $g\,t^{-1}$ Zn. This value is much higher than the estimate of 20.0 $g\,t^{-1}$ Zn for Asian countries in the global inventory (Pacyna and Pacyna, 2002). In 1999, 17 large zinc smelting plants contributed 70% of the total zinc yield. Most of these big plants are located in Liaoning, Guangdong, Gansu, Hunan, and Guangxi Provinces. These large plants emitted 107.9 t of Hg, contributing 73% of the zinc smelting related Hg emissions. Investigation indicated that several big plants used zinc concentrate ore with very high Hg content, some samples even showing Hg content as high as 1000–1500 $g\,t^{-1}$ of zinc concentrate ore (Jiang, 2004). However, due to limited test samples and lack of detailed information on zinc smelting processes in typical Chinese plants, our estimation in this part is subject to high uncertainty.

Table 5
Emission factors for total Hg from other sources

	Source category	Unit	Emission factor
1.	Fuel oil for stationary sources (e.g., power plants, industrial use)	$g\,t^{-1}$ oil	0.014[a]
2.	Gasoline, diesel, and kerosene	$g\,t^{-1}$ oil	0.058[a]
3.	Biofuel combustion	$g\,t^{-1}$ biofuel	0.020[b]
4.	Grassland/savanna burning	$g\,t^{-1}$ grass burning	0.080[c]
5.	Forest burning	$g\,t^{-1}$ forest burning	0.113[d]
6.	Waste and residue burning		
	Agricultural residue	$g\,t^{-1}$ residue	0.037[d]
	Household waste	$g\,t^{-1}$ waste	2.80[e]
7.	Coal mines spontaneous burning	$g\,t^{-1}$ coal	0.02–0.43[f]
8.	Cement production	$g\,t^{-1}$ cement	0.040[g]
9.	Iron and steel production	$g\,t^{-1}$ steel	0.04[h]
10.	Caustic soda production	$g\,t^{-1}$ caustic soda	20.4[i]
11.	Non-ferrous metal smelting		
	Zinc (Zn)	$g\,t^{-1}$ Zn	13.8–156.4[j]
	Copper (Cu)	$g\,t^{-1}$ Cu	9.6[k]
	Lead (Pb)	$g\,t^{-1}$ Pb	43.6[k]
	Gold (Au): large-scale production	$t\,t^{-1}$ Au	0.79[k]
	Gold (Au): artisanal production	$t\,t^{-1}$ Au	15.0[l]
12.	Mercury mining	$kg\,t^{-1}$ Hg	45.0[l]
13.	Battery and fluorescent lamp production	$t\,t^{-1}$ Hg used	0.05[l]

[a]From US EPA (1995).
[b]From Friedli et al. (2003a).
[c]Average emission factor for forests is $0.113\,g\,t^{-1}$ (Friedli et al., 2003b). We assume that grasslands are generally like forests in terms of long-term exposure to Hg, but with typically rather shorter lifetimes for Hg uptake. This value is therefore lowered to $0.080\,g\,t^{-1}$ for grassland burning.
[d]From Friedli et al. (2003b).
[e]From UNECE/EMEP (2004).
[f]Hg content of raw coal as mined for each province is used, and we assume 83% of total Hg is released to atmosphere.
[g]Coal related Hg emissions for cement production are excluded from this category. Energy intensity of $0.196\,t$ of coal $t^{-1}$of cement produced (Zhou et al., 2003) is used here to adjust emission factor of $0.065\,g\,t^{-1}$ of cement (US EPA, 1997) to $0.040\,g\,t^{-1}$ of cement produced.
[h]From Pacyna and Pacyna (2002).
[i]From Qi et al. (2000).
[j]From Jiang (2004). Hg emission factors for zinc smelting at provincial level are used here, with nationally averaged emission factor of $86.6\,g\,t^{-1}$ of zinc produced. Hg emissions from small artisanal zinc smelting in Guizhou Province are also included in this study using data from Feng et al. (2004c).
[k]From Jiang (2004).
[l]From Qi (1997).

Because Hg emissions from gold smelting using amalgamation technology are strongly affected by the size of the smelting plant, we separate the gold smelting process into two parts: large-scale gold smelting in industrial plants and small-scale artisanal gold smelting. Amalgamation technology is gradually being phased out in the large-scale gold smelting plants. In 1999, only about 20 t of gold were produced from large-scale plants using amalgamation technology, which resulted in 16.1 t of total Hg emissions. Although artisanal gold production was small in 1999, Hg emissions were still large due to the high emission factor for this process (see Table 5). We estimate that total Hg emissions from small artisanal gold smelting were 28.5 t in 1999. Artisanal gold smelting was officially banned in China in September 1996, though it persists in remote areas. It is difficult to get precise gold production estimates from these small activities, and our Hg emission estimates from this activity are subject to large uncertainties.

Copper production in China was about 1.1 million tons in 1999. Total Hg emissions from copper smelting were 10.1 t. Hg emissions from copper smelting are much lower than those of zinc smelting due to the use of a lower

Table 6
Speciation of total Hg for each major source type (as fraction of the total)

	Source category	Hg^0	Hg^{2+}	Hg^p
1.	Coal combustion[a]			
	Power plants	0.20	0.78	0.02
	Industrial use			
	(a) w/PM control devices	0.20	0.78	0.02
	(b) Stoker, w/o PM control devices	0.09	0.03	0.88
	(c) Cyclone, w/o PM control devices	0.23	0.35	0.42
	(d) Coke production	0.07	0.58	0.35
	(e) Coke/burned, w PM control devices	0.20	0.78	0.02
	(f) Coke/burned, w/o PM control devices	0.07	0.58	0.35
	Residential and other uses	0.09	0.03	0.88
2.	Fuel oil for stationary sources (e.g., power plants, industrial use)[b]	0.50	0.40	0.10
3.	Gasoline, diesel, and kerosene combustion[b]	0.50	0.40	0.10
4.	Biofuel combustion[c]	0.96	0.00	0.04
5.	Grassland/savanna burning[c]	0.96	0.00	0.04
6.	Forest burning[c]	0.96	0.00	0.04
7.	Waste and residue burning[c]	0.96	0.00	0.04
8.	Coal mines spontaneous burning[d]	0.09	0.03	0.88
9.	Cement production[b]	0.80	0.15	0.05
10.	Iron and steel production[b]	0.80	0.15	0.05
11.	Caustic soda production[b]	0.70	0.30	0.00
12.	Non-ferrous metal smelting[b]	0.80	0.15	0.05
13.	Mercury mining[e]	0.80	0.15	0.05
14.	Battery and fluorescent lamp production[e]	0.80	0.15	0.05

[a]From US EPA (2002).
[b]From Pacyna and Pacyna (2002).
[c]From Friedli et al. (2001, 2003a, b).
[d]Assumed to be the same profile as coal burning for residential use.
[e]Assumed to be the same profile as other non-combustion sources.

emission factor for copper smelting (9.6 g t^{-1} of copper produced) in our study, which mainly results from much lower mercury content in copper concentrate ore than that in zinc concentrate ore. Pacyna and Pacyna (2002) also estimate an emission factor of 10.0 g t^{-1} Cu for copper smelting in Asian countries. Lead production in China was about 0.9 million tons in 1999. We estimate that total Hg emissions from lead smelting were 40.1 t. In 1999, 15 large lead smelting plants contributed 57% of the total lead yield. Most of these large plants are located in Hunan, Yunnan, Henan, and Guangdong Provinces.

3.3. Hg emissions from other source types

Besides the two biggest Hg emitter categories mentioned above, battery and fluorescent lamp production, cement production, mercury mining, and biofuel burning are also major contributors of Hg emissions. Although batteries containing large amounts of Hg are gradually being phased out in China, a significant amount of Hg was consumed in the battery/fluorescent lamp sector in 1999 (see Yang et al., 2003), which resulted in 24.3 t of total Hg emissions. China is the biggest cement producer in the world with 567 million tons of yield in 1999. We estimate that total process Hg emissions in this sector (coal-related emissions are excluded to avoid double-counting with industrial coal use) were 22.7 t in 1999. In China, domestic mercury mining shrunk dramatically in the late 1990s (Hylander, 2001; Hylander and Meili, 2003). The Hg yield in 1999 was down to 195 t, 62% lower than in 1996. We estimate that total Hg emissions from mercury mining were 8.8 t. Most of the mercury mining takes place in Guizhou Province, where the biggest mercury deposit is located. Biofuels dominate rural energy supply in China. Recent work by Friedli et al. (2001, 2003a, b, 2004) and Brunke et al. (2001) has shown that vegetation burning can be a significant source of Hg. In 1999, 413 million tons of biofuels were burned in China, which resulted in 8.3 t of total Hg emissions.

Table 7
Summary of Hg emission estimates (t) associated with fuel consumption and materials production and use in 1999

Source category	Fuel consumption or material yield	Hg	Hg^0	Hg^{2+}	Hg^p
Coal combustion		202.36	33.06	123.89	45.41
(1) Power plants		67.97	13.59	53.02	1.36
(a) Raw coal	487.4×10^6 t[a]	66.18	13.24	51.62	1.32
(b) Cleaned coal	14.5×10^6 t[a]	1.80	0.36	1.40	0.04
(2) Industrial use		103.19	16.66	69.93	16.59
(a) Raw coal	538.2×10^6 t[a]	83.50[j]	14.63	57.78	11.08
(b) Cleaned coal	138.4×10^6 t[a]	17.76[k]	1.67	10.70	5.39
(c) Coke	88.8×10^6 t[a]	1.81	0.33	1.37	0.11
(d) Briquettes	0.9×10^6 t[a]	0.13	0.02	0.09	0.02
(3) Residential use		19.70	1.77	0.59	17.34
(a) Raw coal	105.5×10^6 t[a]	17.23	1.55	0.52	15.16
(b) Cleaned coal	7.8×10^6 t[a]	1.01	0.09	0.03	0.89
(c) Coke	1.4×10^6 t[a]	0.02	0.00	0.00	0.02
(d) Briquettes	8.7×10^6 t[a]	1.44	0.13	0.04	1.26
(4) Other use (e.g., commercial)		11.50	1.03	0.34	10.12
(a) Raw coal	66.2×10^6 t[a]	11.07	1.00	0.33	9.74
(b) Cleaned coal	2.4×10^6 t[a]	0.34	0.03	0.01	0.30
(c) Coke	2.1×10^6 t[a]	0.07	0.01	0.00	0.06
(d) Briquettes	0.1×10^6 t[a]	0.03	0.00	0.00	0.02
Fuel oil for stationary sources	33.8×10^6 t[a]	0.47	0.24	0.19	0.05
Gasoline, diesel, and kerosene	96.8×10^6 t[a]	5.61	2.81	2.25	0.56
Biofuel combustion	413.0×10^6 t[b]	8.26	7.93	0.00	0.33
Grassland/savanna burning	52.1×10^6 t[c]	4.17	4.00	0.00	0.17
Forest burning	25.1×10^6 t[c]	2.83	2.72	0.00	0.11
Waste and residue burning		5.94	5.71	0.00	0.25
Agricultural residue	105.3×10^6 t[c]	3.90	3.74	0.00	0.17
Household waste	0.7×10^6 t[d]	2.05	1.96	0.00	0.08
Coal mines spontaneous burning	30.0×10^6 t[e]	3.02	0.27	0.09	2.66
Cement production	566.9×10^6 t[f]	22.68	18.14	3.40	1.13
Iron and steel production	123.0×10^6 t[f]	4.92	3.94	0.74	0.25
Caustic soda production	9.3×10^3 t[d]	0.19	0.13	0.06	0.00
Non-ferrous metal smelting		242.35	193.88	36.35	12.12
Zinc (Zn)	1.7×10^6 t[g]	147.56	118.05	22.13	7.38
Copper (Cu)	1.1×10^6 t[g]	10.12	8.09	1.52	0.51
Lead (Pb)	0.9×10^6 t[g]	40.08	32.06	6.01	2.00
Gold (Au): large scale	20.4 t[d]	16.10	12.88	2.41	0.80
Gold (Au): artisanal	1.9 t[h]	28.50	22.80	4.28	1.43
Mercury mining	195.0 t[g]	8.78	7.02	1.32	0.44
Battery/fluorescent lamp production	485.0 t[i]	24.25	19.40	3.64	1.21
Total		535.84	299.24	171.92	64.68

[a]From NBS (2001).
[b]From ECCCEY (2000).
[c]From Streets et al. (2003b).
[d]From Jiang (2004).
[e]From Guan et al. (1998).
[f]From NBS (2000).
[g]From ECCNMI (2000).
[h]Artisanal gold smelting activities were officially banned in September 1996, but some mines continue to operate surreptitiously. In our study, we assume artisanal gold production in 1999 is 1.9 t, one-third of 1995 artisanal gold production (Feng, 2005).
[i]This is the amount of Hg used in battery and fluorescent lamp production (Jiang, 2004; Yang et al., 2003).
[j]Hg emissions related to coke production from raw coal are included.
[k]Hg emissions related to coke production from cleaned coal are included.

Other small Hg emission sources include fuel oil (see Wilhelm, 2001); gasoline, diesel and kerosene; grassland and savanna burning; forest burning; agricultural residue and household waste burning; spontaneous burning in coal mines; iron and steel production; and caustic soda production. These sources together emitted 27.2 t of total Hg in 1999. Past production of chlorine and caustic soda by a process that employs flowing elemental mercury once resulted in significant releases of mercury in China, but these plants have now been virtually eliminated (Qi et al., 2000). There are rumored to be factories in China using mercury as a catalyst in the production of polyvinyl chloride (PVC), but we have found no information on such sources.

4. National and regional emission estimates

We estimate that national emissions of total Hg in China in 1999 were 535.8 t, with 299.2 t of Hg^0, 171.9 t of Hg^{2+}, and 64.7 t of Hg^p (Table 7). Our total Hg emissions are apportioned by sector as follows: (1) non-ferrous metals smelting category 45.2%, in which the sector splits are 27.5% from zinc smelting, 8.3% from gold smelting, 7.5% from lead smelting, and 1.9% from copper smelting; (2) coal combustion category 37.8%, in which the sector splits are 19.3% from industrial use, 12.7% from power plants, 3.7% from residential use, and 2.1% from other uses; (3) battery and fluorescent lamp production 4.5%; (4) cement production 4.2%; (5) mercury mining 1.6%; (6) biofuels 1.5%; and (7) all other small sectors together 5.1%. The biggest three single sectors are zinc smelting, industrial coal use, and coal-fired power plants, contributing 59.5% of total Hg emissions in China in 1999. Hg^0 is the largest component species, contributing 55.8% of total Hg emissions, followed by Hg^{2+} 32.1% and Hg^p 12.1%.

Table 8 shows the summary of emission estimates by province. In 1999, the three highest emitting provinces are Liaoning (54.1 t or 10.1% of the national total), Guangdong (44.2 t), and Guizhou (39.0 t). The reason that Liaoning and Guangdong are high emitters is attributed to the presence of non-ferrous metal smelters, e.g., large zinc smelting plants in Liaoning, and large zinc and lead smelting plants in Guangdong. For Guizhou, the reason is due to the high Hg content of raw coal in this province and the relatively large amount of uncontrolled coal combustion that takes place.

Figs. 1a–c shows the distributions of total Hg emissions in China in 1999 at a resolution of 30×30 min from all sources combined, the coal combustion category alone, and the non-ferrous metals smelting category alone, respectively. Figs. 2a–c presents the distributions for each Hg species (Hg^0, Hg^{2+}, and Hg^p, respectively). In total, 283 power plants (73% of total power plant emissions), 128 industrial plants (6% of total emissions related to industrial coal use), 17 zinc smelting plants (73% of total zinc smelting emissions), 15 lead smelting plants (58% of total lead smelting emissions), and 15 household waste burning plants (100% of emissions in this sector) were treated as point sources. Their emissions were precisely located at their latitude/longitude coordinates. The surplus emissions (total minus point sources) were distributed using various spatial proxies at 1×1 km resolution (see Streets et al., 2003a; Woo et al., 2003). The provincial emissions from Table 8 were used as control totals.

Figs. 1b and c clearly show that the distribution of total Hg emissions from coal combustion is much different than that from non-ferrous metals smelting. Hg emissions from the latter category are primarily scattered in the south/southwest of China (Guangdong, Guangxi, Yunnan, Hunan, and Guizhou Provinces), in some provinces in the Yellow River area (Gansu, Shaanxi, and Henan Provinces), and in Liaoning Province. However, Hg emissions from coal combustion are concentrated around the populated and industrial centers of China—the coastal provinces in east and north China. Besides these coastal provinces, Guizhou Province stands out in coal-related Hg emissions due to the high Hg content of raw coal in this province and the relatively large amount of coal burned without particulate control devices. Total Hg emissions from all sources together, as shown in Fig. 1a, reflect the combined characteristics of these two biggest Hg emitters (coal combustion and non-ferrous metal smelting sources), as well as the distribution for each Hg species (Hg^0, Hg^2, and Hg^p), as shown in Figs. 2a–c. As a result, Hg emissions from all sources together are much more evenly distributed than those from coal combustion alone or non-ferrous metals smelting alone.

5. Uncertainties

Quantifying Hg emissions is more difficult than quantifying, say, SO_2 or NO_x, because the emissions

Table 8
Summary of Hg emission estimates (t) by province in 1999

Province	Coal combustion	Non-ferrous metals smelting	Other sources	Total Hg	Hg^0	Hg^{2+}	Hg^p
Anhui	10.01	1.39	3.15	14.56	5.49	7.29	1.78
Beijing	5.11	2.02	1.70	8.83	3.80	3.12	1.92
Fujian	1.88	0.02	1.76	3.66	1.73	1.56	0.37
Gansu	1.25	26.06	1.49	28.80	22.17	4.80	1.83
Guangdong	7.42	31.10	5.72	44.24	30.82	10.94	2.48
Guangxi	5.37	22.18	2.54	30.09	20.80	7.13	2.16
Guizhou	19.24	9.57	10.20	39.00	18.39	11.08	9.53
Hainan	0.24	0.00	0.56	0.79	0.51	0.23	0.06
Hebei	13.29	4.66	4.51	22.46	9.40	9.35	3.71
Heilongjiang	4.49	1.40	3.35	9.24	4.82	3.62	0.80
Henan	14.03	16.09	4.67	34.80	18.88	11.64	4.27
Hong Kong	0.81	0.00	1.28	2.08	0.97	1.00	0.12
Hubei	8.76	3.74	3.45	15.95	7.29	6.46	2.20
Hunan	3.64	25.49	3.21	32.34	23.63	6.43	2.27
Jiangsu	11.78	3.16	4.52	19.46	8.35	9.37	1.74
Jiangxi	3.90	3.64	2.10	9.64	5.31	3.32	1.00
Jilin	5.73	1.79	1.71	9.23	3.70	3.85	1.67
Liaoning	10.28	40.66	3.12	54.06	36.45	12.76	4.85
Macao	0.00	0.00	0.01	0.01	0.00	0.00	0.00
Nei Mongol	8.56	4.06	3.69	16.31	7.42	5.99	2.90
Ningxia	1.41	0.00	0.57	1.98	0.52	0.93	0.53
Qinghai	0.21	0.99	0.85	2.06	1.58	0.29	0.19
Shaanxi	2.52	9.75	1.69	13.96	9.46	3.17	1.33
Shandong	13.03	1.02	6.07	20.12	7.75	9.44	2.92
Shanghai	6.14	1.07	1.38	8.60	2.91	4.67	1.01
Shanxi	15.32	0.64	1.83	17.79	3.68	9.31	4.81
Sichuan	9.40	6.06	6.10	21.56	11.51	7.51	2.53
Tianjin	3.33	0.24	0.82	4.40	1.34	2.13	0.93
Xinjiang	0.44	4.89	1.91	7.24	5.52	1.11	0.61
Xizang	0.00	0.00	1.20	1.20	1.14	0.01	0.05
Yunnan	6.79	17.91	2.60	27.30	17.50	6.73	3.06
Zhejiang	8.00	2.73	3.38	14.10	6.38	6.70	1.02
Total	202.36	242.35	91.13	535.84	299.24	171.92	64.68

come from so many source types, not primarily combustion sources. In this respect Hg emissions are similar to VOC emissions. We acknowledge that for some types of sources very little is known about actual activity levels and emission factors, and our choices in such cases rely heavily on inferences of activity levels from quite limited and uncertain statistical information. On the other hand, at least for combustion sources and releases from mercury-containing ores, total emissions are constrained by the Hg content of the coal or raw material, in a similar way to the sulfur content of fossil fuels, and this acts to reduce the uncertainty.

Several factors influence the estimation of emissions, including emission factor and activity level. We estimate the uncertainty for each emitting sector by combining the coefficients of variation (CV, or the standard deviation divided by the mean) of the contributing factors. We then combine these uncertainties to estimate the total uncertainty of Hg emission estimates by quadrature average when the source estimates are uncorrelated. We follow the same detailed methodology for uncertainty analysis that was described in the TRACE-P inventory paper of Streets et al. (2003a). Fig. 3 shows the results of uncertainty estimation in Hg emissions by source type. Our general findings are that Hg emissions are known least well in the artisanal gold smelting sector ($\pm 450\%$), followed by the mercury mining sector ($\pm 340\%$). Emissions are best known in the coal combustion category: $\pm 40\%$ for power plants, $\pm 60\%$ for industrial coal use, and $\pm 70\%$ for

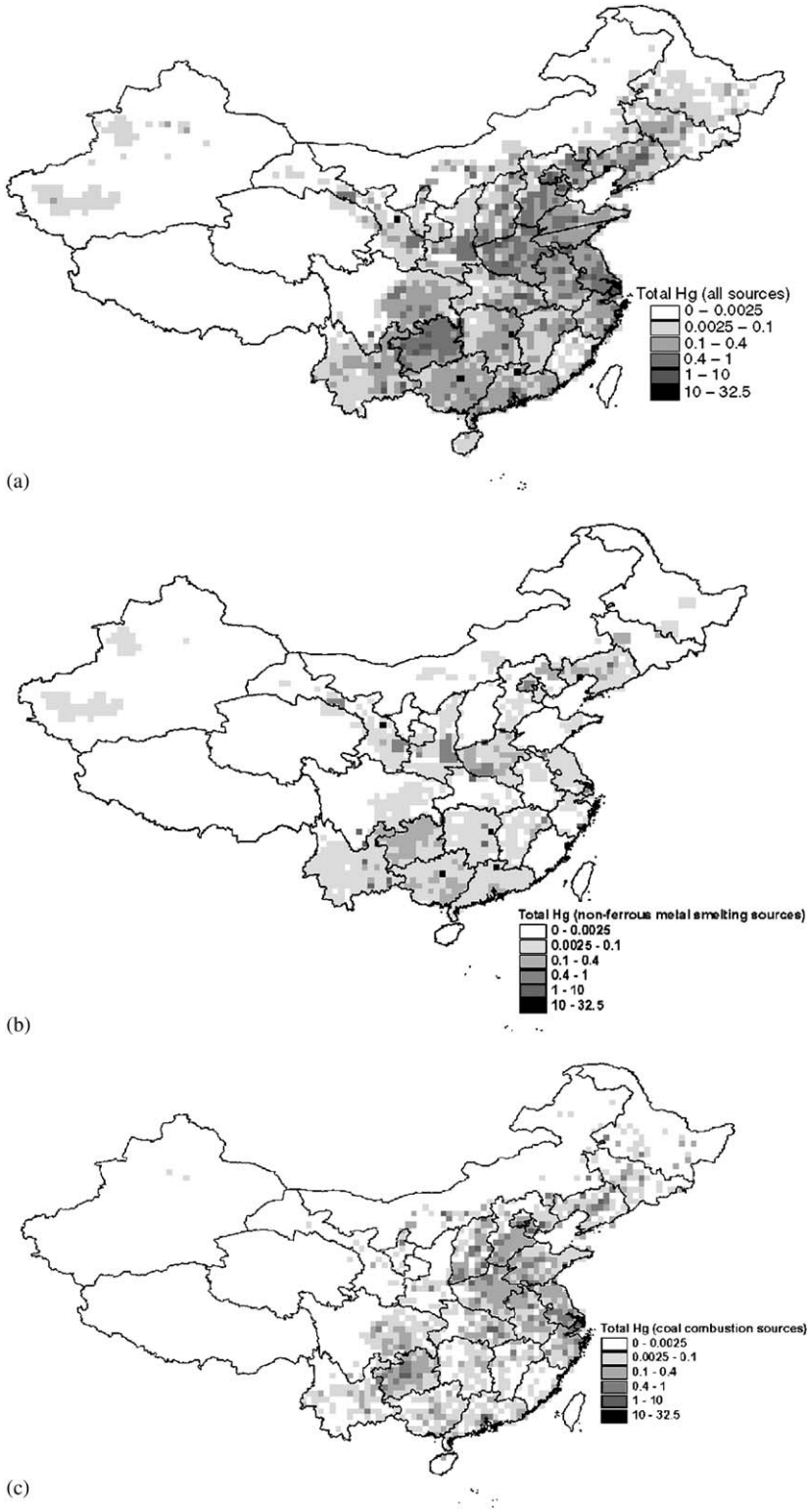

Fig. 1. (a) Gridded total Hg emissions for the year 1999 (all sources together, 30×30 min resolution, unit: t yr^{-1} per grid cell). (b) Gridded total Hg emissions for the year 1999 (coal combustion category alone, 30×30 min resolution, unit: t yr^{-1} per grid cell). (c) Gridded total Hg emissions for the year 1999 (non-ferrous metals smelting category alone, 30×30 min resolution, unit: t yr^{-1} per grid cell).

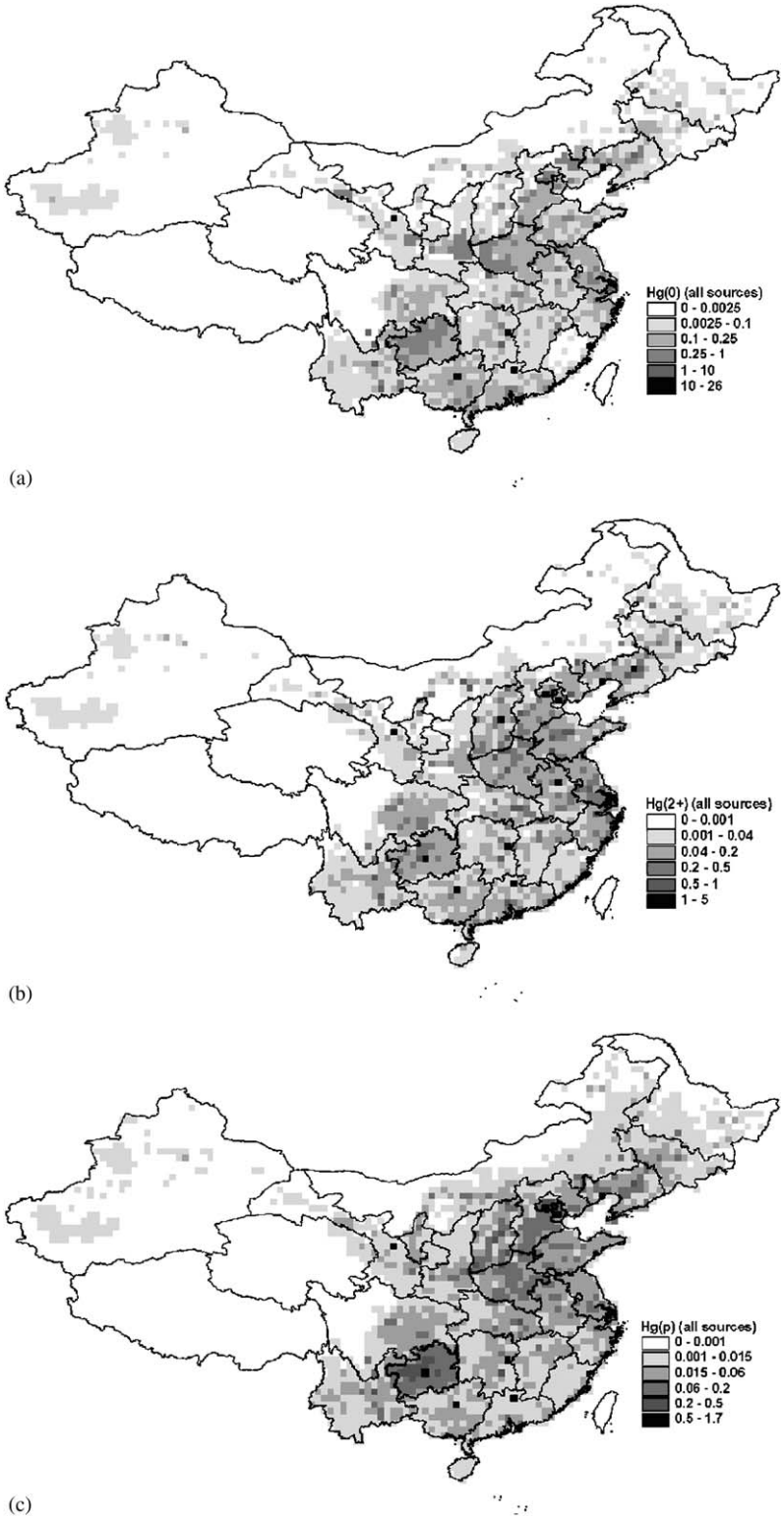

Fig. 2. (a) Gridded Hg^0 emissions for the year 1999 (all sources combined, 30×30 min resolution, unit: t yr^{-1} per grid cell). (b) Gridded Hg^{2+} emissions for the year 1999 (all sources combined, 30×30 min resolution, unit: t yr^{-1} per grid cell). (c) Gridded Hg^p emissions for the year 1999 (all sources combined, 30×30 min resolution, unit: t yr^{-1} per grid cell).

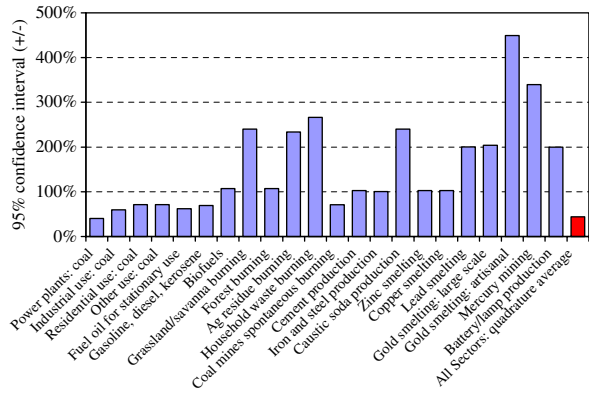

Fig. 3. Uncertainty (%) in Hg emission estimates by sector (95% confidence intervals, ±).

residential coal use and other uses. The overall uncertainty in emissions for all sectors combined is estimated to be ±44%. This value can be compared with estimates of ±13% for China's SO_2 emissions, ±23% for NO_x emissions, ±59% for VOC emissions, and ±156% for CO emissions (Streets et al., 2003a). As the confidence intervals are frequently greater than the mean, our presentation of relative confidence intervals $>\pm 100\%$ might suggest that the lower confidence interval is negative. However, the true confidence interval is not symmetric about the mean because some the underlying variables are lognormally distributed. A better interpretation of "±400%", for example, might be "within a factor of five" so that the confidence interval would be 20–500% of the mean given.

6. Conclusions and discussion

We present for the first time a detailed estimation of China's mercury emissions by province, gridded for use in atmospheric models. Thirty-seven contributing source types are examined, and the analysis for coal combustion considers the province-specific distribution of technology types. Our estimate is that China's emissions in 1999 were 536 (±236) t. These are direct anthropogenic inputs to air, excluding natural sources and re-emission. This level of primary emissions is rather similar to the estimation by Pacyna and Pacyna (2002)—fortuitously, because the source category contributions are different. Our value for coal combustion, 202 t in 1999, is similar to the Chinese estimate of 214 t in 1995. Approximately 45% of the Hg comes from non-ferrous metals smelting, 38% from coal combustion, and 17% from miscellaneous activities, of which battery and fluorescent lamp production and cement production are the largest. We find that emissions are concentrated in southwestern China (particularly Guizhou Province), the east central and coastal regions of industrialized and populated China, and Liaoning Province in the northeast.

We estimate that 56% of the Hg in China is released as Hg^0, 32% as Hg^{2+}, and 12% as Hg^p. These are not too dissimilar to the global values of 53%, 37%, and 10%, respectively, cited by Pacyna and Pacyna (2002). Our general expectation would be (a) that Hg^p emissions would be higher in China than in the rest of the world due to the heavy burning of coal in low-temperature residential and industrial settings without PM controls; (b) that emissions of Hg^{2+} from coal-fired power plants in China would be relatively higher than Hg^0 due to the absence of flue-gas desulfurization units, which tend to dissolve the soluble Hg^{2+}; and (c) that the releases of Hg from metals smelting would be heavily favoring elemental mercury. Inter-regional comparisons of speciation profiles are needed to improve the transcontinental modeling of atmospheric Hg transport.

Several studies have attempted to infer China's mercury emission strength by analysis of measurements off the Chinese coast, coupled with regional atmospheric chemistry modeling. Friedli et al. (2004) analyzed ACE-Asia aircraft observations taken in the western Pacific Ocean during spring 2001 and estimated that Hg emissions from China were about 750 t yr^{-1}. Jaffe et al. (2005) inferred emissions of 1460 t yr^{-1} by analysis of ground-station measurements taken at Okinawa island during spring 2004. Pan et al. (2005) concluded from a modeling analysis of ACE-Asia observations that emissions in China are underestimated by 60–100%; because their initial assumed inventory value was 510 t, this means they infer that emissions were ~800–1000 t yr^{-1} in 2001.

These values are consistent with our primary source term estimate of 536 t for 1999. First, we know from work in progress that Hg emissions in China have grown since 1999, especially in the period 2003–2004, due to continued increases in coal consumption and industrial production. Our preliminary estimates are 575 t for 2001 and ~750 t for 2004. Second, we might expect that re-emission in China would be large due to extensive deposition of Hg to soil and water bodies during prior years of

heavy coal burning. Feng et al. (2005) estimate that mercury emission fluxes from soil (presumably re-emission plus natural) in Guiyang, Guizhou Province, are about 64% of primary emissions. Applying this factor to our primary source estimates for the whole of China—for illustration purposes only—yields values of 940 t for 2001 and 1230 t for 2004. Third, a number of sources are omitted from this comparison, including non-Chinese anthropogenic sources (Korea, Japan, etc.), volcanic emissions, and emissions from transpiration of vegetation (a potentially important unknown).

Many uncertainties remain in our knowledge of primary anthropogenic releases of mercury to the atmosphere in China. Because many of the activities that release large amounts of mercury occur in remote parts of the country (and may actually be illegal), they tend to lie on the fringe of official statistics. Residential coal use in rural areas may be under-reported in Chinese statistics. We are lacking in actual measurements of Hg emissions from Chinese combustors and the capture of Hg in Chinese emission control devices. There are even large discrepancies in estimates of the typical Hg content of coal in many provinces. Until these shortcomings can be remedied by a program of field testing, uncertainty in the Hg emissions estimate will persist.

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