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Trends in Anthropogenic Mercury Emissions in China from 1995 to 2003

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We have developed multiple-year inventories of anthropogenic mercury emissions in China for 1995 through 2003. We estimate that total Hg emissions from all anthropogenic sources increased at an average annual rate of 2.9% during the period 1995−2003, reaching 696 (±307) t in 2003, with a speciation split of 395 t of Hg^0, 230 t of Hg^{2+}, and 70 t of Hg^p. Nonferrous metals smelting and coal combustion continue to be the two leading mercury sources in China, as nonferrous metals production and coal consumption keep increasing. Nonferrous metals smelting and coal combustion together contributed ∼80% of total Hg emissions during the past decade. Hg emissions from coal combustion increased from 202 t in 1995 to 257 t in 2003 at an average annual rate of 3.0%. Among all of the coal consumption sectors, the power sector is the leading one in Hg emissions growth, up by 5.9% annually. Hg emissions from nonferrous metals smelting increased from 230 t in 1995 to 321 t in 2003 at an average annual rate of 4.2%. Although Hg emissions related to gold smelting decreased since 1996, other nonferrous metals such as zinc, lead, and copper contributed significant Hg growth at annual rates of 8.5%, 13.0%, and 6.9%, respectively. At provincial level, the trends of Hg emissions show significant variation. The uncertainty level decreased from ±78% (95% confidence interval) in the estimate of total emissions in 1995, to ±44% in 2003. This is primarily attributed to the decreased emissions from those Hg sources with the largest uncertainty in both activity levels and emission factors, such as artisanal gold smelting, mercury mining, and battery/fluorescent lamp production.

1. Introduction

During the past decade, worldwide environmental mercury (Hg) levels have increased considerably, and accordingly, 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 (*1*, *2*). In March 2005, the U.S. Environmental Protection Agency (EPA) led the way in regulating mercury by issuing the Clean Air Mercury Rule (CAMR) to permanently cap and reduce mercury emissions from coal-fired power plants (*3*). In June 2005, the Canada-Wide Standards for Mercury were also released in draft form (*4*).

Mercury contamination is a serious problem in China. Feng (*5*) has summarized a number of specific instances associated with industrial releases of Hg in past years. High concentrations of Hg in the air of China's cities have been reported in several studies (*6*−*10*). However, studies on Hg emissions in China are quite limited. Wang et al. (*11*) estimated that 214 t of Hg were released from coal combustion in 1995. Pacyna and Pacyna (*12*) did not report calculated emissions from China per se but estimated that Asian emissions in 1995 were 860 t from stationary fuel combustion and 1074 t in total and suggested that China's emissions were approximately 500 t. The assessments of global source attribution for mercury by Seigneur et al. (*13*, *14*) estimated that Asian anthropogenic Hg emissions in 1998 were 1117 t, in which the Chinese data were simply extrapolated from the 1990 gridded global emission inventory of the Global Emission Inventory Activity (GEIA) and Wang's 1995 data. This previous body of work is inadequate because it is either incomplete or lacking in local knowledge of Hg sources.

Since 2003, Argonne National Laboratory and Tsinghua University have been developing a comprehensive inventory of mercury emissions from anthropogenic sources in China, following the precedent of the Asian TRACE-P emission inventory (*15*, *16*). We developed 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 (no natural sources or re-emission). Hg emissions are speciated using technology-specific factors and gridded for use in atmospheric models. A detailed estimation of China's mercury emissions by province for the year of 1999 is presented in Streets et al. (*17*).

However, an inventory for a single year may not be enough to show the current mercury status in China, since China's energy consumption has increased significantly during the past decade. This is especially important because the year of our detailed study, 1999, is coincidentally located at the bottom of a temporary energy stagnation period (1997−2000). In this study, multiple-year inventories of anthropogenic mercury emissions in China for 1995 through 2003 are presented. The methodology is consistent with the previous detailed analysis of 1999 emissions. The trends of Hg emissions by sector and by province are discussed in detail.

2. Methodology, Data Sources, and Key Assumptions

Mercury emissions are calculated using fuel consumption data and detailed Hg emission factors. The basic concept of the Hg emission calculation is described by the following equation:

$$E_t = \sum_i \sum_j [\mathrm{ef}_{i,j,t} A_{i,j,t} F_{\mathrm{REL}j,t} (1 - F_{\mathrm{REM}j,t})] \qquad (1)$$

where E_t is the Hg emission; $\mathrm{ef}_{i,j,t}$ is the Hg content of coal as burned or emission factor for other fuels or noncombustion processes; $A_{i,j,t}$ is the amount of fuel consumption or production yield of noncombustion processes; $F_{\mathrm{REL}j,t}$ is the fraction of Hg released to the atmosphere; $F_{\mathrm{REM}j,t}$ is the fraction of Hg removed by emission control devices; j is the combustor

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TABLE 1. Hg Content of Raw Coal as Mined in China, by Province (mg kg^{-1})

province[a]	Hg content	province[a]	Hg content
Anhui	0.26	Jiangxi	0.22
Beijing	0.44	Jilin	0.20
Fujian	0.08	Liaoning	0.17
Gansu	0.05	Nei Mongol	0.22
Guangdong	0.15	Ningxia	0.20
Guangxi	0.30	Qinghai	0.04
Guizhou	0.52	Shaanxi	0.11
Hainan	0.15	Shandong	0.18
Hebei	0.14	Shanxi	0.16
Heilongjiang	0.09	Sichuan	0.14
Henan	0.25	Xinjiang	0.02
Hubei	0.16	Yunnan	0.29
Hunan	0.10	Zhejiang	0.35
Jiangsu	0.16		
	National Average by Year		
1995	0.180	2000	0.187
1996	0.183	2001	0.185
1997	0.186	2002	0.183
1998	0.189	2003	0.186
1999	0.185		

[a] Hong Kong, Macao, Shanghai, Tianjin, and Xizang do not produce raw coal and are not included in this table.

type with/without emission control devices; i is the province; and t is the year.

2.1. Mercury Content of Coal and Coal Products. Table 1shows the provincial Hg content of raw coal, as mined in China. The values used in our study are merged from USGS data and Chinese literature data (*17*). We assume that the provincial-level Hg content of raw coal as produced did not change during the time period 1995−2003. The national average Hg content by production (see Table 1) varies slightly over time, 0.18−0.19 mg kg^{-1}, due to fluctuations in provincial coal production.

The methodologies for calculating the Hg content of cleaned coal, coal briquettes, and coke as produced are documented in our previous paper (*17*). We assume an average coal cleaning Hg removal efficiency of 30% that is independent of the Hg content, and that 10% of the Hg contained in a given coal remains in 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.

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 to relate the coal produced (mined) in particular provinces to its consumption in each province. As a result, a transportation matrix is required to quantify in-province coal use and inter-province coal flows. In this study, only 1996 and 1999 coal transportation matrixes were set up due to statistical data restrictions (*17*−*22*). We found that the inter-province coal supply patterns were relatively steady for the majority of provinces between 1996 and 1999, resulting in a small change (<10%) in Hg content in coal as burned for most of the provinces. Therefore, in this study, we apply the 1996 transportation matrix to 1995, 1997 and 1998, and the 1999 matrix to the remaining years (2000 to 2003). Then the Hg content of coal for the consuming provinces is calculated as follows:

$$\mathrm{ef}_{c,i,j,t} = M_{i,j,t} ef_{s,i,j,t} \quad (2)$$

where $\mathrm{ef}_{c,i,j,t}$ is the Hg content of coal in the consuming provinces; $M_{i,j,t}$ is the transportation matrix; $\mathrm{ef}_{s,i,j,t}$ is the Hg content of coal in the supplying provinces; i is the coal type (raw coal, cleaned coal, coke, briquette); j is the sector (power, coking, industrial, residential, other uses); and t is the year. A detailed comparison of the Hg content of raw coal as produced and as burned in China by province for 1999 can be found in our previous paper (*17*).

TABLE 2. Hg Removal Efficiencies of FGD and Various PM Control Devices (%)

	removal efficiency	refs
ESP+FGD	74.0	25
ESP	30.6	17
PM scrubber	6.5	17
cyclone	0.1	17

2.2. Coal Consumption, Combustor Types and Emission Controls by Sector. Multiple-year coal consumption data by sector and coal type are provincial-level estimates compiled from the China Energy Statistical Yearbooks (*21*−*24*). Total raw coal consumption increased from 1.46 billion tons in 1995 to 1.96 billion tons in 2003, at an average annual rate of 3.7% nationwide. Among the major coal consuming sectors, the power sector is the leading sector in total coal growth, increasing 7.2% annually. The industrial sector has a moderate increase in total coal use, 2.4% annually. Total coal use from the residential sector has been slightly decreasing (−1.1% annual rate) due to fuel transitions to cleaner gaseous fuels in many regions of China. Similarly, coal consumption for other uses (e.g., transportation, construction, and others) has been slightly decreasing, by about −1.2% annually, again mainly due to fuel transitions.

Because the Hg release rates and the speciation profiles depend greatly on combustion technology and conditions, it is necessary to define Chinese coal utilization practices. Our model contains 65 individual source types for coal combustion, 22 of which are for coal-fired power plants, 30 for industrial use, nine for residential use, and four for other uses. The partitioning of each combustion technology/control device/fuel type by province and sector over time is built into our model based on a wide literature review (*17*).

In the past decade, the installation of PM control devices in boilers increased significantly in China, especially in the power sector. Since the mid-1980s, electrostatic precipitators (ESP) increased their share by 4−5% annually, to replace wet particle scrubbers and cyclones in power plants. Now the share of ESP installation in the total coal-fired power capacity is close to ∼95% nationwide. However, in the industrial sector, the penetration of PM control installation lags far behind. Although installation of wet particle scrubbers increased steadily over the past decade, the fraction of industrial coal use without any PM control device is still large at present, close to 30%. The reasons are (1) a large number of small boilers scattered throughout China, especially in the poorer and more remote provinces, are without PM control and (2) coke ovens, consuming a large amount of raw coal and clean coal, are generally without PM control. Since the mid 1990s, flue-gas desulfurization (FGD) began to be installed in power plants to reduce SO_2 emissions in China. In 1995, FGD installated capacity was only 0.7 GW; however, by the end of 2003, the FGD capacity had reached 6.9 GW, ∼2.5% of total coal-fired generating capacity, mostly in Sichuan (including Chongqing), Beijing, Shandong, Guangdong, Heilongjiang, Jiangsu, and Zhejiang Provinces. In this study, we applied these time-varying provincial-level technology data for our emission inventory calculations.

The shares of Hg remaining in the bottom ash for different boiler types in China are documented in our previous paper (*17*). Table 2 presents assumed Hg removal efficiencies of the three predominant types of PM control devices and FGD

TABLE 3. Summary of Total Hg Emission Estimates (t) by Sector, 1995−2003

source category	1995	1996	1997	1998	1999	2000	2001	2002	2003	AAGR[a]
coal combustion	202.4	209.3	208.2	207.6	202.2	204.3	208.8	225.5	256.7	3.0%
(1) power plants	63.4	68.7	67.2	66.2	67.8	70.1	76.3	84.2	100.1	5.9%
(2) industrial use	104.7	106.3	107.8	108.3	103.2	104.2	101.9	109.9	124.3	2.2%
(3) residential use	23.1	23.5	22.7	21.5	19.7	19.6	19.9	19.7	21.7	−0.8%
(4) other uses	11.2	10.8	10.5	11.6	11.5	10.5	10.7	11.8	10.6	−0.7%
nonferrous metals smelting	230.1	213.1	212.2	213.8	242.4	262.4	281.7	294.6	320.5	4.2%
(1) zinc (Zn)	97.6	103.2	125.5	127.8	147.6	161.4	173.0	178.5	187.6	8.5%
(2) copper (Cu)	10.4	10.7	11.3	8.4	10.1	12.7	13.7	14.8	17.6	6.9%
(3) lead (Pb)	26.5	30.8	30.9	33.0	40.1	48.0	54.3	57.8	70.7	13.0%
(4) gold (Au): large scale	10.1	11.4	16.1	16.1	16.1	11.8	12.3	15.0	16.2	6.0%
(5) gold (Au): artisanal	85.5	57.0	28.5	28.5	28.5	28.5	28.5	28.5	28.5	−12.8%
fuel oil for stationary sources	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.6	2.3%
gasoline, diesel, and kerosene	4.3	4.6	4.6	5.0	5.6	6.1	6.4	6.8	7.6	7.2%
biofuel combustion	10.1	9.1	8.7	8.7	8.3	8.6	9.5	10.6	10.7	0.7%
grassland/savanna burning[b]	4.2	4.2	4.2	4.2	4.2	4.2	4.2	4.2	4.2	0.0%
forest burning[b]	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.8	0.0%
agricultural residue burning[b]	3.9	3.9	3.9	3.9	3.9	3.9	3.9	3.9	3.9	0.0%
household waste burning	0.6	0.6	0.6	2.0	2.0	2.8	3.2	7.7	10.4	42.5%
coal mines spontaneous burning[b]	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0	0.0%
cement production	19.9	20.5	21.3	21.4	22.7	23.9	27.0	29.4	35.0	7.4%
iron and steel production	3.8	4.1	4.4	4.6	4.9	5.1	6.1	7.3	8.9	11.2%
caustic soda production	2.4	2.4	2.5	1.3	0.2	0.2	0.2	0.2	0.0	N/A
mercury mining	35.1	22.9	37.6	10.1	8.8	9.1	8.7	22.3	27.5	−3.0%
battery/fluorescent lamp production	29.1	34.1	49.7	37.6	24.5	16.2	8.7	6.2	3.7	−22.7%
total	**552.2**	**534.9**	**564.2**	**526.4**	**535.7**	**553.0**	**574.7**	**625.0**	**695.6**	**2.9%**
(a) Hg^0	**311.7**	**293.4**	**317.4**	**288.0**	**299.2**	**312.2**	**327.1**	**357.7**	**394.9**	**3.0%**
(b) Hg^{2+}	**169.2**	**171.3**	**176.5**	**170.8**	**171.8**	**177.9**	**184.8**	**202.0**	**230.3**	**3.9%**
(c) Hg^p	**71.4**	**70.2**	**70.3**	**67.6**	**64.7**	**62.9**	**62.8**	**65.3**	**70.3**	**−0.2%**

[a] Annual average growth rate. [b] Assumed no change over time due to lack of data.

installed in boilers in China. We assume that these values do not change over time.

2.3. Other Fuel Consumption and Material Yields. Besides coal combustion, there are many other combustion sources (other fossil fuels, biofuels, etc.) and noncombustion sources (cement production, ferrous and nonferrous metals smelting processes, etc.) contributing to Hg emissions in China. Multiple-year liquid-fuel (e.g., gasoline, diesel, etc.) and biofuel consumption data are provincial-level estimates compiled from the China Energy Statistical Yearbooks (*21*−*24*). Material-yield data (e.g., cement, nonferrous metals) related to noncombustion processes by province are from the China Statistical Yearbooks (*26*) and professional yearbooks (*27*−*30*) such as the Yearbooks of Nonferrous Metals Industry of China.

Total liquid-fuel consumption increased from 100.2 million tons in 1995 to 158.6 million tons in 2003, at an annual rate of 5.9% nationwide. Among the major oil fuels, the total consumption of gasoline, diesel, and kerosene contributed the major increment. Biofuel consumption initially decreased from 506.2 million tons in 1995 to 413 million tons in 1999, then increased again to 536.7 million tons in 2003. The production of cement, iron and steel, and nonferrous metals each increased significantly in China during the past decade, at an annual growth rate of more than 7.0%. China has become the largest producer of cement and iron and steel. Artisanal gold smelting was officially banned in China in September 1996; however, some of these activities still persist in remote areas. It is difficult to get precise gold production estimates for these small-scale activities. We assume that the artisanal gold yield in 1996 was two-thirds of the level in 1995, and that from 1997 to 2003 the yield was one-third of the level in 1995.

2.4. Hg Emission Factors for Other Sources. Emission factors for other sources are documented in our previous paper (*17*). Hg emission factors for spontaneous burning in coal mines and for zinc smelting are available at provincial level; however, only nationally averaged emission factors are available for most of the other source types due to lack of local data. We assume these emission factor values do not change during the period 1995−2003.

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). The speciation of Hg compounds for each Hg emission source sector is detailed in our previous paper (*17*). We assume these values do not change during the period 1995−2003.

3. Results and Discussion

3.1. Emission Trends by Sector. Total Hg emissions by sector from 1995 to 2003 are summarized in Table 3. We estimate that national emissions of total Hg in China in 1995 were 552.2 t, increasing to 695.6 t in 2003, at an annual average growth rate of 2.9%. As coal consumption and nonferrous metals yields keep increasing, coal combustion and nonferrous metals smelting continue to be the two leading mercury sources in China. Of the major source contributions to total anthropogenic Hg emissions shown in Figure 1, coal combustion contributed nearly 40% of total, and nonferrous metals smelting contributed ∼40 to ∼45%. These two categories comprise the majority of China's anthropogenic Hg emissions, ∼80% of total, and their shares kept relatively constant during the past decade.

Mercury emissions from coal combustion increased from 202.4 t in 1995 to 256.7 t in 2003 at an annual average rate of 3.0%. Among all of the coal consuming sectors, Hg emissions from the power sector are increasing fastest, 5.9% annually and reaching 100.1 t in 2003. We estimate that about 42 t of Hg were removed in the power sector in 2003 based on current ESP installation capacity; however, this did not offset the increase of Hg emissions in this sector due to growth in electricity production. As ESP is now close to its maximum Hg reduction potential in China, penetration of new and more efficient control technologies, such as FGD and activated carbon injection (ACI), is necessary to achieve more Hg reduction benefits in the future. Fortunately, the China

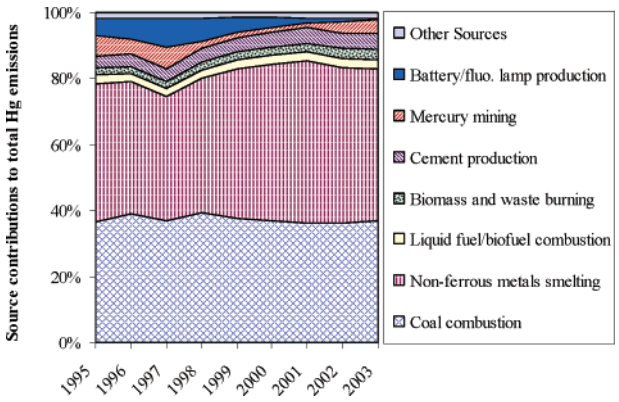

FIGURE 1. Source contributions to total Hg emissions in China, 1995−2003.

State Council approved the State Environmental Protection Administration (SEPA) *Control Plan for Acid Rain and SO_2 Pollution Control Zone* in 1998, and the penetration of FGD in the power sector is expected to increase significantly in the next decade. Although total Hg emissions from the industrial sector did not increase as fast as from the power sector, industrial coal combustion is still the largest single sector for Hg emissions among all the coal consumption sectors, increasing from 104.7 t in 1995 to 124.3 t in 2003. We estimate that about 18 t of Hg were removed by coal washing in 2003, and most of this reduction accrued to the industrial sector since the industrial sector consumed nearly 90% of all cleaned coal. Coal consumption for residential and other uses decreased during the period due to fuel transitions, resulting in Hg emission decreases (see Table 3). In 2003, total Hg emissions from residential use and other uses were 21.7 and 10.6 t, respectively.

Nonferrous metals smelting operations are known to be another large source of mercury in China (*17*). Due to fast economic growth in China, the demand for nonferrous metals is increasing significantly. As a result, total Hg emissions from the nonferrous metals smelting category (zinc, lead, copper, and gold) increased rapidly at an annual rate of 4.2%, from 230.1 t in 1995 to 320.5 t in 2003. In this category, zinc smelting is the largest single sector in total Hg emissions, reaching 187.6 t in 2003 at an average annual rate of 8.5%. However, lead smelting was the leading sector in Hg emissions growth: from 26.5 t in 1995 to 70.7 t in 2003, increasing by 13.0% annually. Total Hg emissions from copper smelting increased to 17.6 t in 2003, at an annual rate of 6.9%. Gold smelting is the only sector with decreased Hg emissions in this category, attributed to an official ban of small artisanal gold production in China in 1996. In 2003, total Hg emissions from gold smelting were 44.7 t, compared to 95.6 t in 1995. It should be noted that our estimate of emissions from metals smelting is subject to high uncertainty due to limited test samples, lack of detailed information on metal smelting processes in typical Chinese plants, and lack of precise production estimates from small activities.

Besides the two biggest Hg emitter categories mentioned above, cement production, mercury mining, battery and fluorescent lamp production, household waste burning, and biofuel burning are also major contributors of Hg emissions during the whole period or part of the period (1995−2003). Total Hg emissions from cement production (coal-related emissions are excluded to avoid double-counting with industrial coal use) increased steadily from 19.9 t in 1995 to 35.0 t in 2003, at an annual rate of 7.4%. In China, domestic mercury mining shrunk dramatically in the late 1990s, but has rebounded since 2002. As a result, the total Hg emissions from mercury mining fluctuated significantly, decreasing from 35.1 t in 1995 to 8.7 t in 2001, then back up to 27.5 t in 2003. Hg-containing batteries are being phased out in China due to the release of a stringent standard in December 1997. Therefore, total Hg emissions from this sector increased initially, from 29.1 t in 1995 to 49.7 t in 1997, then decreased significantly since 1998, to as low as 3.7 t in 2003. Biofuels dominate rural energy supply in China. Total Hg emissions from biofuel burning remain nearly constant, around 10 t annually. Although household waste burning contributed only 0.6 t of Hg emissions in 1995, it is the leading sector among all of the Hg source sectors in emission growth, reaching 10.4 t in 2003 at an annual growth rate over 40%. Among other miscellaneous small sources in this study (see Table 3), liquid fuels (gasoline, diesel, and kerosene) and iron and steel production are two sectors with high Hg growth, at annual average growth rates of 7.2 and 11.2%, respectively.

We estimate that 57% of the Hg in China in 2003 is released as Hg^0, 33% as Hg^{2+}, and 10% as Hg^p, compared to 56% as Hg^0, 31% as Hg^{2+}, and 13% as Hg^p in 1995. A slight decrease in Hg^p emissions is primarily attributed to Hg emission reductions from residential coal use and other coal use, artisanal gold mining, and battery production. The increases of Hg^0 and Hg^{2+} emissions are primarily driven by zinc and lead smelting and coal-fired power plants. As the ability of Hg^0 to be transported over long distances has been confirmed by both modeling studies (*13*, *31*, *32*) and observation (*33*, *34*), China may contribute more to the regional/global Hg transportation cycle if the trend of increased Hg^0 emissions during the past decade keeps up.

3.2. Regional Emission Trends. Total Hg emissions by province for 1995 and 2003 are summarized in Table 4. At the provincial level, the trends of total Hg emissions show significant differences. Some provinces (e.g., Ningxia and Guangdong Provinces) show much higher Hg emission growth over the national average during the past decade; however, some other provinces (e.g., Xinjiang and Heilongjiang Provinces) show reduced Hg emissions over this period. Hg emissions for each province are strongly affected by specific source-related trends. For example, (1) the large increase in mercury emissions, 12.8% annually, for Ningxia Province is primarily attributed to greatly increased coal consumption in the industrial sector; (2) the 7.9% increase in annual mercury emissions in Guangdong Province is driven by increased zinc smelting and large-scale promotion of household waste burning; (3) Hg emissions reductions in Beijing (−0.2% annually) are primarily due to fast penetration of control technologies, such as ESP and FGD, in the power sector; and (4) Hg emissions reductions were achieved in Liaoning Province (−1.7%) by cutting zinc and lead production. As a result, the different regional trends in Hg emissions may result in different trends in Hg concentrations in the air at the local/regional scale. For example, at multiple-year scale, we may be able to see increasing Hg concentrations in the air in East China and South China, but relatively constant or even slightly decreasing Hg concentrations in the air in the metropolitan area of Beijing/Tianjin.

For a specific Hg emission source type, the Hg emission trends can be quite different from one province to another. Figures 2a and b present two examples: annual growth rates of total Hg emissions in the power sector and the residential sector at provincial level, respectively. As shown in Figure 2a, Fujian Province, the leading province in power-related Hg emissions growth, has as high as ∼14% annual average growth rate for the period. For comparison, Guangxi, Ningxia, and Heilongjiang Provinces exhibit annual Hg emission growth rates of less than 2% from 1995 to 2003. It should be noted that Beijing had reduced Hg emissions in the power sector in the past decade, which is attributed to much faster penetration of control technologies, such as ESP and FGD.

As mentioned above, coal use from the residential sector has been decreasing due to fuel transitions in many regions

TABLE 4. Summary of Total Hg Emission Estimates (t) by Province, 1995 and 2003

province	1995	2003	AAGR[a]
North China			
Beijing	6.3	6.2	−0.2%
Tianjin	6.0	5.3	−1.4%
Hebei	36.7	28.4	−3.1%
Shanxi	19.3	26.4	4.0%
Nei Mongol	16.3	16.8	0.4%
Northeast China			
Liaoning	52.8	45.9	−1.7%
Jilin	13.6	10.0	−3.8%
Heilongjiang	11.5	8.3	−4.0%
East China			
Shanghai	7.9	10.5	3.7%
Anhui	15.6	23.6	5.3%
Jiangsu	20.8	23.6	1.6%
Zhejiang	12.9	21.0	6.3%
Fujian	3.8	5.4	4.5%
Jiangxi	12.0	14.8	2.6%
Shandong	21.9	27.1	2.7%
Central China			
Henan	41.8	59.1	4.4%
Hubei	14.2	17.6	2.7%
Hunan	26.2	47.2	7.6%
South China			
Guangdong	26.4	48.5	7.9%
Guangxi	24.0	35.1	4.8%
Hainan	0.5	0.8	6.2%
Hong Kong	2.4	2.5	0.6%
Macao	0.01	0.01	4.5%
Southwest China			
Guizhou	34.8	55.5	6.0%
Sichuan	23.8	37.5	5.8%
Yunnan	33.1	35.8	1.0%
Xizang	2.7	1.2	−9.9%
West China			
Gansu	15.2	28.7	8.3%
Ningxia	2.2	5.7	12.8%
Qinghai	1.9	1.9	0.0%
Shaanxi	31.8	38.8	2.5%
Xinjiang	13.7	6.1	−9.6%
Total	**552.2**	**695.6**	**2.9%**

[a] Annual average growth rate.

in China, resulting in general decreases in Hg emissions. Figure 2b shows this trend. Nearly two-thirds of the provinces have decreases in Hg emissions in the residential sector. For example, Hg emissions in the residential sector in Guangdong Province decreased by more than 15% annually since 1995. However, some regions, such as Nei Mongol and Ningxia Provinces, still have positive increases (~7% annually) in Hg emissions; this means that the fuel transition in these areas could not offset the increasing demand for coal.

3.3. Uncertainties. Quantifying Hg emissions is more difficult than quantifying, say, SO_2 or NO_x, because the emissions 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 (e.g., artisanal gold smelting) 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 methodology for uncertainty analysis that was described in detail in the TRACE-P inventory paper of Streets et al. (*15*). The uncertainty estimates for individual emitting sectors were discussed in our previous paper (*17*). Figure 3 shows the results of uncertainty estimation in the Hg emissions trend from 1995 to 2003. In general, the uncertainty level has diminished over time. In 1995 we calculate an uncertainty level of ±78% (95% confidence interval) in the estimate of total emissions, or ±429 tons. Since then, the uncertainty level has become smaller. This is primarily attributed to decreased emissions from those Hg sources that have the largest uncertainty in both activity levels and emission factors, such as artisanal gold smelting, mercury mining, and battery/fluorescent lamp production. In 2003, the uncertainty level is reduced to ±44% of total emissions, or ±307 tons. These uncertainty values 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 (*15*).

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. (*33*) 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}. Pan et al. (*35*) concluded from a modeling analysis of ACE-Asia observations that emissions in China are

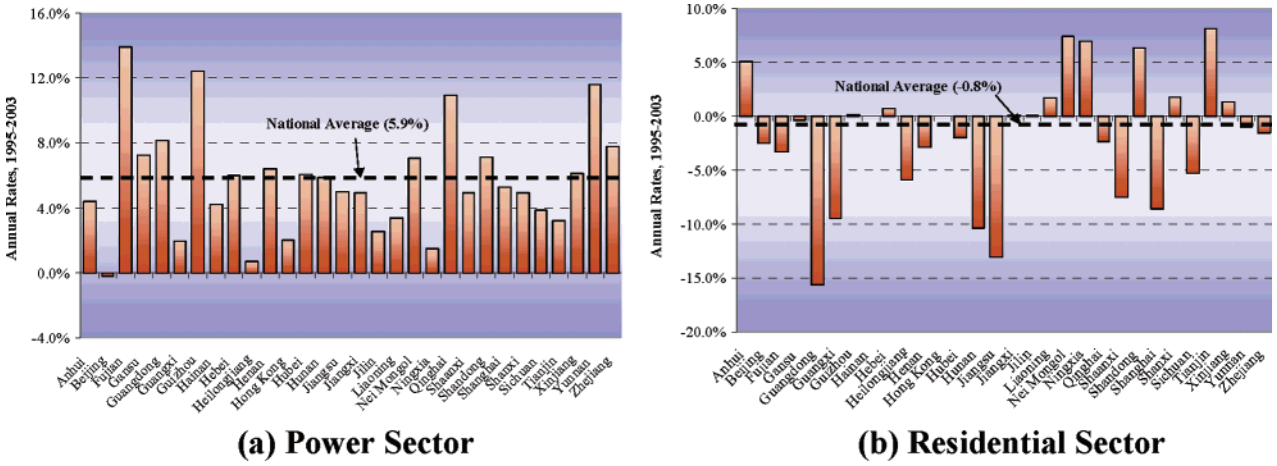

FIGURE 2. Annual rates of Hg emissions growth at provincial level, 1995−2003.

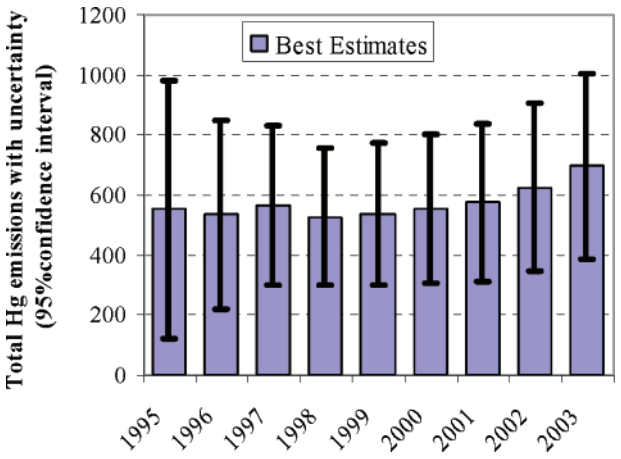

FIGURE 3. Uncertainty in total Hg emissions from 1995 to 2003 (95% confidence interval).

underestimated by 80−200%; because their initial assumed inventory value was 510 t, this means they infer that emissions were ∼900−1500 t yr^{-1} for China in 2001. Jaffe et al. (*34*) inferred Hg^0 emissions of 1460 t yr^{-1} from Asia by analysis of ground-station measurements taken at Okinawa, Japan, during spring 2004. China emissions are estimated to be ∼50% of the total Hg emissions in Asia by several studies (*12*, *35*) and the fraction of Hg^0/total Hg is ∼60% (*12*, *17*). If we apply these two factors to Jaffe's estimate for the total Hg emissions of China, for illustration purposes only, it yields a value of ∼1200 t for 2004.

These values are consistent with our primary source term estimate of 575 ± 261 t for 2001 and 696 ± 307 t for 2003. First, Our preliminary estimate for 2004 further increased to ∼765 ± 337 t (∼10% increase from 2003). 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 and metal smelting. Pan et al. (*35*) estimate that re-emissions over land, biogenic and marine sources represent about 80% of primary emissions in Asia. Feng et al. (*36*) estimate that mercury emission fluxes from soil (presumably re-emission plus natural) in Guiyang, Guizhou Province, China, are about 64% of primary emissions. If we simply apply these factors for illustration purposes only, re-emissions in China could be as large as ∼370−460 t for 2001 and ∼490−610 t for 2004, respectively. And, it should be noted, re-emission is subject to much greater uncertainty than primary emissions due to poor knowledge on this area at present. 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). When the man-made and natural source terms are combined, they may well explain the observed fluxes. This is a topic for refinement in future research.

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 actual measurements of Hg emission rates and Hg species profiles from Chinese combustors/smelters and the capture of Hg in Chinese emission control devices. There are even large discrepancies in estimates of the typical Hg content of coal and concentrate ore 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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