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Updated Emission Inventories for Speciated Atmospheric Mercury from Anthropogenic Sources in China

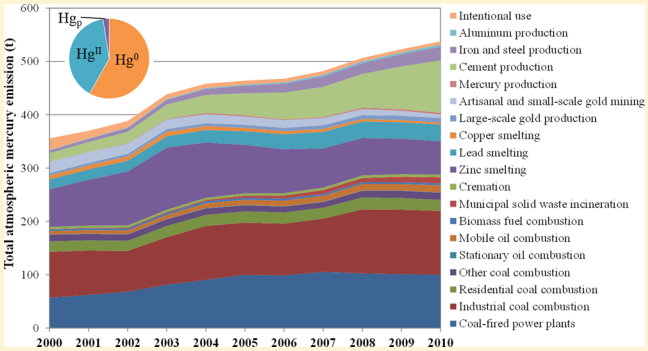
Lei Zhang,[†,§] Shuxiao Wang,*[,†,‡,§] Long Wang,[†] Ye Wu,[†,‡] Lei Duan,[†,‡] Qingru Wu,[†] Fengyang Wang,[†] Mei Yang,[†] Hai Yang,[†] Jiming Hao,[†,‡] and Xiang Liu[†]

[†]School of Environment, and State Key Joint Laboratory of Environment Simulation and Pollution Control, Tsinghua University, Beijing 100084, China

[‡]State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing 100084, China

Supporting Information

ABSTRACT: China is the largest contributor to global atmospheric mercury (Hg), and accurate emission inventories in China are needed to reduce large gaps existing in global Hg mass balance estimates and assess Hg effects on various ecosystems. The China Atmospheric Mercury Emission (CAME) model was developed in this study using probabilistic emission factors generated from abundant on-site measurements and literature data. Using this model, total anthropogenic Hg emissions were estimated to be continuously increasing from 356 t in 2000 to 538 t in 2010 with an average annual increase rate of 4.2%. Industrial coal combustion, coal-fired power plants, nonferrous metal smelting, and cement production were identified to be the dominant Hg emission sources in China. The ten largest contributing provinces accounted for nearly 60% of the total Hg emissions in 2010. Speciated Hg emission inventory was developed over China with a grid-resolution of 36 × 36 km, providing needed emission fields for Hg transport models. In this new inventory, the sectoral Hg speciation profiles were significantly improved based on the latest data from field measurements and more detailed technology categorization. The overall uncertainties of the newly developed inventory were estimated to be in the range of −20% to +23%.



1. INTRODUCTION

Mercury (Hg) has drawn global attention due to its persistence, toxicity, long-range transport, and bioaccumulation in the environment. Atmospheric mercury is divided into three chemical forms, including gaseous elemental mercury (GEM or Hg^0) and reactive gaseous mercury (RGM or Hg^{II}) and particle-bound mercury (PBM or Hg_p).[1] Over 90% of the total atmospheric mercury is Hg^0 with a residence time of several months to a year in the lower atmosphere.[2] The first legally binding international treaty aimed at controlling and reducing global Hg emissions, Minamata Convention on Mercury, was approved at the fifth session of the Intergovernmental Negotiating Committee on mercury (INC5) in January, 2013. China is the largest emitter of atmospheric mercury in the world.[3] To better evaluate the environmental impact of anthropogenic Hg emissions in China, accurate emission inventories for China are of great importance.

Streets et al.[4] (2005) developed the first complete anthropogenic Hg emission inventory of China and estimated the total Hg emission in China to be 536 t in 1999. A cooperative study[5] between Tsinghua University and Argonne National Laboratory analyzed the historical trend of Hg emissions from anthropogenic sources in China from 1995 to 2003 and found an average annual increasing rate of 2.9% during this period. The total anthropogenic Hg emission in China reached 696 t in 2003. The Hg emission inventory dedicated for anthropogenic sources in China was only updated to 2003. The recent anthropogenic Hg emission inventories were evaluated in the global level. Pacyna et al.[6] (2010) estimated that Hg emissions of China in 2005 reached 825 t, accounting for 43% of the global emissions. Pirrone et al.[7] (2010) estimated that China emitted 609 t of mercury in 2007, and stationary combustion and nonferrous metal production accounted for 44% and 33% of the national emissions, respectively. In the 2013 technical report of global mercury assessment by AMAP and UNEP,[8] the Hg emissions from anthropogenic sources in China were estimated to be 575 t in 2010. The comparison between different studies is shown in Table S1 in the Supporting Information.

The predominant Hg emission sources in China's inventory are coal combustion, nonferrous metal smelting, cement production, and iron and steel production. Jiang et al.[9] (2005) developed a detailed Hg emission inventory for the

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coal combustion sector in China and got a total emission at 162 and 220 t in 2000 with two sets of coal mercury data, respectively. A historical trend of Hg emission from coal combustion during 1980−2007 was developed by Tian et al.[10] (2010), and total emission from coal combustion was estimated to be 306 t in 2007. Hylander and Herbert[11] (2008) calculated the Hg emissions from zinc, lead, and copper smelting in China to be totally 83.2 t. On the basis of recent field measurement results, Wu et al.[12] (2012) updated the Hg emission inventory for zinc, lead, and copper smelting to be 72.5 t in 2010. With onsite measurements in several cement plants, Li, in his thesis[13] (2011), assessed the total Hg emission from cement production in China in 2008 to be 20.6 t. Yang[14] (2014) collected and analyzed limestone samples from all over China, conducted field tests in cement plants, updated the Hg emission inventory for cement production in China, and found the total Hg emission from cement production to be 93.5 t in 2010. The study of Tian et al.[15] (2012) showed that the total Hg emissions from municipal solid waste (MSW) incineration in China increased from 5.35 t in 2003 to 36.7 t in 2010, whereas Hu et al.[16] (2012) and Chen et al.[17] (2013) also estimated the Hg emissions from MSW incineration in 2010 and got 6.1 and 4.7 t, respectively. Zhang et al.[18] (2013) developed Hg emission inventories for biomass fuel combustion in China and found the total emission in the range of 2.9−3.8 t from 2000 to 2007. It can be seen that there are large differences among previous inventories due to both emission factor selection and estimating methods. Therefore, it is difficult to examine temporal trends without consistency in methods used to estimate emissions.

Large uncertainties exist in the existing inventories. Wu et al.[5] (2006) used a semiquantitative approach based on uncertainty ranking of each parameter in inventory development and estimated the results of their study had the uncertainty level of ±78% and ±44% (95% confidence interval) in 1995 and 2003, respectively. The main cause of the inventory uncertainty lies in the emission factors for major Hg emission sources, such as coal combustion, nonferrous metal smelting, and cement production.[19] With the results of field measurements accumulating and the understanding of emission mechanism deepening, the Hg emission factors for the major sectors became more and more accurate, and researchers updated the inventories for major Hg emission sectors in China with new experimental yield. However, with respect to Hg emissions from either all anthropogenic sources or major emission sources, a deterministic emission factor approach was employed in most previous studies. This method has its limitation and is unable to reflect the probabilistic distribution characteristics of the key factors in the model.

The purpose of this study is to obtain the temporal and spatial distributions of atmospheric Hg emissions from anthropogenic sources in China for the period of 2000−2010. A novel methodology, that is, probabilistic technology-based method according to the information on Hg contents of fuel/raw materials, production process, and Hg removal efficiencies obtained from field tests, was used in this study to acquire more accurate emission estimates and lower uncertainties. Activity levels were refined, and parameters for the Hg emission factors were updated with the latest data from field measurements. The inventories yielded from this study will give a clearer picture on the change of historical Hg emission in China from 2000 to 2010 to quantify the cobenefit of air pollution control strategies in China on mercury removal, and provide spatial distribution of different Hg species in China to serve air quality models.

2. DATA AND METHODOLOGY

2.1. Model description. The model developed in this study for estimating speciated anthropogenic atmospheric Hg emissions in China was named China Atmospheric Mercury Emission (CAME) model. Hg emission sources in this model were divided into 6 categories and 23 subcategories, shown in Table S2 in the Supporting Information. The conventional model is deterministic emission factor model, which uses average values for the input parameters in emission estimation. The core of the CAME model is probabilistic technology-based emission factor, described as eq 1:

$$E(x, y, z) = \sum_i A_i \cdot M_i(x) \cdot (1 - f_i \cdot w(y)) \cdot \sum_j \sum_k R_j \cdot (1 - P_{jk} \cdot \eta_{jk}(z)) \quad (1)$$

where $E(x,y,z)$ is the probabilistic distribution of the amount of Hg emission from a certain sector; i is the province; j is the type of boiler or technique; k is the type of air pollution control device (APCD) combination; A is the activity level of a given emission sector; $M(x)$ is the probabilistic distribution of the mercury concentration in fuel/raw material; f is the fraction of the pretreatment of fuel or raw material; $w(y)$ is the probabilistic distribution of the mercury removal rate by the pretreatment; R is the mercury release rate of a certain type of boiler or technique; P is the proportion of a certain type of APCD combination; $\eta(z)$ is the probabilistic distribution of the mercury removal rate by a certain type of APCD combination.

The CAME model incorporates Monte Carlo simulations to take into account the probabilistic distributions of key input parameters and produce Hg emission results in the form of a statistical distribution. As discussed in detail in the following section, core input parameters such as mercury concentration in fuel/raw material and mercury removal efficiency by APCDs fit the skewed distribution (e.g., log-normal distribution and Weibull distribution). Therefore, the arithmetic mean values used in deterministic models were not able to reflect the best guesses of these key parameters, which could probably result in overestimation or in rare cases underestimation of the Hg emissions from given sectors. The calculations in the CAME model for the dominant sectors, coal combustion, nonferrous metal smelting, and cement production (accounting for over 80% of the inventory) were all technology-based and the APCD categorization was more detailed and updated. All the improvements of methodology in this study contributed to more accurate inventories.

Based on dominance of the Hg emission sources and the availability of essential data, all the subcategories were ranked into four tiers (see Table S2 in the Supporting Information), each tier using a different calculation method. Estimates for emissions from Tier 1 sectors are from previous detailed studies.[18,20] Sources in Tier 2 use overall emission factors due to shortage of on-site measurements in these sources. Emission factors for Tier 2 emission sources are listed in Table S3 in the Supporting Information. Emission sources in Tier 3 and Tier 4 cover the most important sources in China, including coal combustion, nonferrous metal smelting, and cement production, responsible for over 80% of the total anthropogenic emissions in China. Estimates for sources in Tier 3 use probabilistic technology-based emission factor model based on the updated database of the Hg concentration in fuel/raw

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Table 1. Mercury Removal Efficiency by Typical APCD Combinations (%)[a]

emission source	APCD combination	mean	min	max	SD	number of tests	probabilistic distribution	reference
coal combustion	WS	23	7	59	18	8	Weibull	Zhang et al.[21] (2012) Wang et al.[22](2010) Zhang[23] (2012)
	ESP	29	1	83	19	64	Weibull	
	FF	67	9	92	30	10	Weibull	
	ESP+WFGD	62	13	88	22	19	Weibull	
	FF+WFGD	86	77	97	10	3	normal	
	SCR+ESP+WFGD	69	36	95	24	4	normal	
	SCR+FF+WFGD	93	86	99	9	2	normal	
	ESP+CFB-FGD+FF	68	68	68		1	normal	
nonferrous metal smelting	DC+FGS+ESD+DCDA	97.8	94.8	99.7	2.0	6	normal	Wu et al.[12] (2012) Wang et al.[25] (2010) Li et al.[27] (2010) Zhang et al.[24] (2012)
	DC+FGS+ESD+MRT+DCDA	99.2	99.1	99.3	0.2	2	normal	
	DC+FGS+ESD+SCSA	86.5	86.5	86.5		1	normal	
	DC+FGS	41	27	55	20	4	normal	
	DC	12	2	20	7	4	normal	
	FGS	33	17	49	23	2	normal	
cement production	shaft kiln/rotary kiln technology without dust recycling	62	57	67	7	2	normal	Li[13] (2011) Wang et al.[26] (2014)
	dry-process precalciner technology with dust recycling	4.1	1.9	6.1	2.1	3	normal	

[a]WS, wet scrubber; ESP, electrostatic precipitator; FF, fabric filter; WFGD, wet flue gas desulfurization; CFB-FGD, circulating fluidized bed flue gas desulfurization; SCR, selective catalytic reduction; DC, dust collector; FGS, flue gas scrubber; ESD, electrostatic demister; MRT, mercury reclaiming tower; DCDA, double conversion double absorption; SCSA, single conversion single absorption.

material and the Hg removal efficiency of APCDs. Estimates for sources in Tier 4, i.e., coal-fired power plants, use a coal-quality-based probabilistic emission factor model, taking into account the influence of coal quality on Hg removal efficiency of APCDs, details of which can be found in our previous paper.[21] Methods for the four tiers are described in detail in Section 1 of the Supporting Information.

2.2. Key Parameters for Different Tiers of Emission Sources. The Hg emission sources in Tier 3 and Tier 4 are dominant ones in China, and their key parameters are discussed in detail as follows.

2.2.1. Mercury Concentration in Fuel/Raw Material. Mercury concentration data of 494 raw coal samples, 381 zinc concentrate samples, 198 lead concentrate samples, 207 copper concentrate samples, and 167 limestone samples (for cement production) were collected in our previous studies.[12,14,21] Interprovincial transport matrix for coal was derived from our previous studies to convert mercury concentration in coal as mined into mercury concentration in coal as consumed, and so was the transport matrix for metal concentrate.[12,21] Imports of coals and nonferrous metal concentrates are also considered in the transport matrix. With the batch fit function of the software Crystal Ball, the mercury concentrations in coal, metal concentrates and limestone were all found to fit the log-normal distributions. Detailed data is listed as Table S4 in the Supporting Information.

2.2.2. Mercury Removal Efficiency of APCDs. Onsite measurement results were investigated from existing literature for mercury removal efficiencies of APCDs for coal combustion, nonferrous metal smelting, and cement production, as summarized in Table 1. Typical APCD combinations for coal combustion include ESP, ESP+WFGD, FF, WS, and FF+WFGD (all the acronyms for APCDs are defined in the notes of Table 1). Previous studies[21−23] contributed 104 onsite measurements for the coal combustion sector. Crystal Ball was applied for batch fit, and mercury removal efficiencies of APCD combinations for nonpower coal combustion were found to fit the Weibull distribution. Onsite measurements for nonferrous metal smelters and cement plants[12,13,24−27] provided data for typical APCD combinations in smelters and cement plants. However, the numbers of tests were not large enough to perform a distribution fitting. Mercury removal efficiencies of APCDs in these sources were assumed to fit normal distributions. The new dry-process precalciner technology with dust recycling in cement production has much lower mercury removal efficiency than shaft kiln/rotary kiln technology.[13,26]

2.2.3. Mercury Speciation in Exhausted Flue Gas. The ultimate mercury speciation profiles in exhausted flue gas from different anthropogenic emission sources are crucial to environmental impacts on local and regional scales in China. Mercury speciation data from existing literature[12,13,21−39] was reviewed and summarized. Hg^0 is the dominant Hg species for most APCD combinations for coal combustion except FF, because WFGD and WS remove a large amount of Hg^{II} in flue gas. A typical nonferrous metal smelter with DCDA process inside acid plant has similar percentages of Hg^0 and Hg^{II}. Mercury reclaiming tower (MRT) can remove a large portion of Hg^0, resulting in high proportion of Hg^{II} in flue gas. Cement production using dry-process precalciner technology with dust recycling and typical iron and steel production both have high Hg^{II} in exhausted flue gas. These updated profiles are quite different from the previous studies[4,5] and will probably affect the Hg speciation profiles of the whole anthropogenic emission inventory of China. Details are shown in Table S5 in the Supporting Information.

2.2.4. Activity Levels and APCD Installation Rates. The activity level of the combustion process is the amount of fuel consumed, and that of the production process is the amount of product. Activity levels of Hg emission sources in China from 2000 to 2010 were derived from official statistics.[40−42] It should be noted that the activity level of nonferrous metal smelting

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Table 2. Summary of Total Mercury Emissions (t) in China by Sector, 2000−2010

emission sector	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	AAGR[a]
coal combustion	175.1	176.6	175.6	203.9	224.5	230.3	227.9	236.4	257.6	257.4	253.8	3.8
coal-fired power plants	57.0	62.0	68.5	81.4	90.6	99.7	98.9	105.4	102.5	101.0	100.0	5.8
industrial coal combustion	86.0	83.3	76.3	89.0	100.9	97.9	96.9	99.8	120.1	121.0	119.7	3.4
residential coal combustion	19.6	19.2	18.8	21.1	20.7	21.3	21.0	20.5	22.2	22.1	20.7	0.5
other coal combustion	12.5	12.0	12.0	12.4	12.3	11.4	11.0	10.7	12.7	13.4	13.5	0.8
other combustion	14.6	15.6	16.8	18.0	19.9	22.3	25.1	27.0	28.7	31.1	33.9	8.8
stationary oil combustion	0.5	0.5	0.5	0.6	0.7	0.6	0.6	0.6	0.5	0.4	0.5	−0.3
mobile oil combustion	6.5	6.7	7.2	7.8	9.1	9.8	10.6	11.2	12.2	12.4	13.5	7.6
biomass fuel combustion	2.9	3.2	3.6	3.6	3.7	3.7	3.8	3.5	3.5	3.5	3.5	2.0
municipal solid waste incineration	0.7	1.1	1.5	1.8	2.2	4.0	5.7	7.2	7.8	10.1	11.6	33.1
cremation	4.1	4.1	4.1	4.1	4.2	4.2	4.5	4.6	4.7	4.7	4.8	1.6
nonferrous metal smelting	96.4	113.1	129.7	146.4	134.6	122.9	116.5	110.2	105.9	101.7	97.4	0.1
zinc smelting	70.6	85.9	101.3	116.6	103.8	90.9	82.3	73.7	70.1	66.5	62.9	−1.1
lead smelting	18.2	19.2	20.2	21.2	23.3	25.5	28.2	30.9	30.9	31.0	31.0	5.5
copper smelting	7.6	7.9	8.3	8.6	7.5	6.4	6.0	5.5	4.9	4.2	3.5	−7.4
precious metal production	26.0	24.9	24.7	24.3	24.6	23.5	22.0	22.0	21.1	20.3	18.8	−3.2
large-scale gold production	4.3	4.5	4.7	5.0	5.3	5.6	6.1	6.9	7.2	8.0	8.8	7.3
artisanal and small-scale gold mining	21.2	20.0	19.0	18.1	17.0	15.7	14.4	13.5	11.3	9.4	6.8	−10.7
mercury production	0.4	0.4	1.0	1.2	2.3	2.2	1.5	1.6	2.7	2.9	3.2	22.8
building material production	21.9	25.6	29.7	37.1	46.7	57.8	69.3	79.8	87.1	106.4	128.1	19.3
cement production	16.0	18.7	21.2	26.7	33.7	41.6	50.0	56.9	63.4	80.1	98.3	19.9
iron and steel production	5.1	6.1	7.3	8.9	11.3	14.1	16.8	19.6	20.1	22.9	25.5	17.4
aluminum production	0.7	0.9	1.1	1.5	1.8	2.1	2.5	3.3	3.5	3.4	4.3	19.2
intentional use	21.8	14.3	11.8	8.9	8.0	7.4	7.0	6.7	6.4	6.1	5.8	−12.4
chlor-alkali production	2.8	2.8	2.8	2.6	2.6	2.6	2.6	2.6	2.6	2.6	2.6	−0.7
reagent production	1.9	1.9	1.9	1.9	1.9	1.9	1.9	1.9	1.9	1.9	1.9	0.0
thermometer production	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.0
fluorescent lamp production	13.4	7.3	5.2	3.0	2.3	1.8	1.5	1.2	1.0	0.7	0.5	−28.0
battery production	3.0	1.6	1.2	0.7	0.5	0.4	0.3	0.3	0.2	0.2	0.1	−27.8
total anthropogenic emission	355.7	370.0	388.2	438.5	458.2	464.1	467.7	482.1	506.7	523.0	537.8	4.2

[a]Annual average growth rate (%).

was converted from the amount of metal product to the amount of metal concentrate consumed based on the grade of the concentrate and the metal recovery rate, and the activity level of cement production was converted from the amount of clinker to the amount of limestone used based on the limestone/clinker ratio. The installation rates of APCD combinations for coal combustion and nonferrous metal smelting was from the previous studies[12,43−45] of our research group. The national application rate of the new dry-process precalciner technology in cement production increased from 10% in 2000 to 82% in 2010, and the rest proportion was shaft kiln/rotary kiln technology. Activity levels for 2000 to 2010 and APCD application rates for 2000, 2005, and 2010 are listed in Table S6 and Table S7 in the Supporting Information, respectively.

2.3. Approach for Uncertainty Analysis. Streets et al.[4] (2005) analyzed the uncertainties of Hg emission inventory of anthropogenic sources in China with a semiquantitative approach by grading all the parameters in Hg emission factors. It was only applicable to the deterministic emission factor model and had relatively lower reliability than the quantitative method. Wu et al.[19] (2010) used the P10−P90 confidence interval of the statistical distribution of Hg emission as the uncertainty range, where P10/P90 value represents a probability of 10%/90% that the actual result would be equal to or below the P10/P90 value. However, this uncertainty range was even larger than the result from the study of Streets et al.,[4] because these two methods were not comparable. Therefore, a new approach was developed here to determine the uncertainty range of a general skewed distribution for the CAME model. The calculating method is shown as eq 2:

$$u^{\pm} = \frac{\mathrm{Mo} - \sqrt{\sigma_s^{\pm}\sigma_k^{\pm}}}{\mathrm{P50}} - 1 \tag{2}$$

where u is the uncertainty; Mo is the mode value; P50 is the value at which there is a probability of 50% that the actual result would be equal to or below; σ_s^- and σ_s^+ are the distances between Mo and the values where the probability equal to $f(\mathrm{Mo})/2$; σ_k^- and σ_k^+ are the distances between Mo and P20 or P80.

The uncertainty range of a normal distribution is described by the relative standard deviation (RSD). The approach for uncertainty analysis in this study extended the use of the RSD in a normal distribution case to a general skewed distribution case. The uncertainty range yielded from this quantitative approach reflects both the span and the kurtosis of the skewed distribution, which is more reasonable and distinguished from a confidence interval. This approach is better to compare with previous studies, e.g., Streets et al.[4] (2005). P10/P90 ranges from the study of Wu et al.[19] (2010) can be better referred as the confidence interval with a confidence degree of 80%. More details can be found in the "Methodology for uncertainty analysis" section in the Supporting Information.

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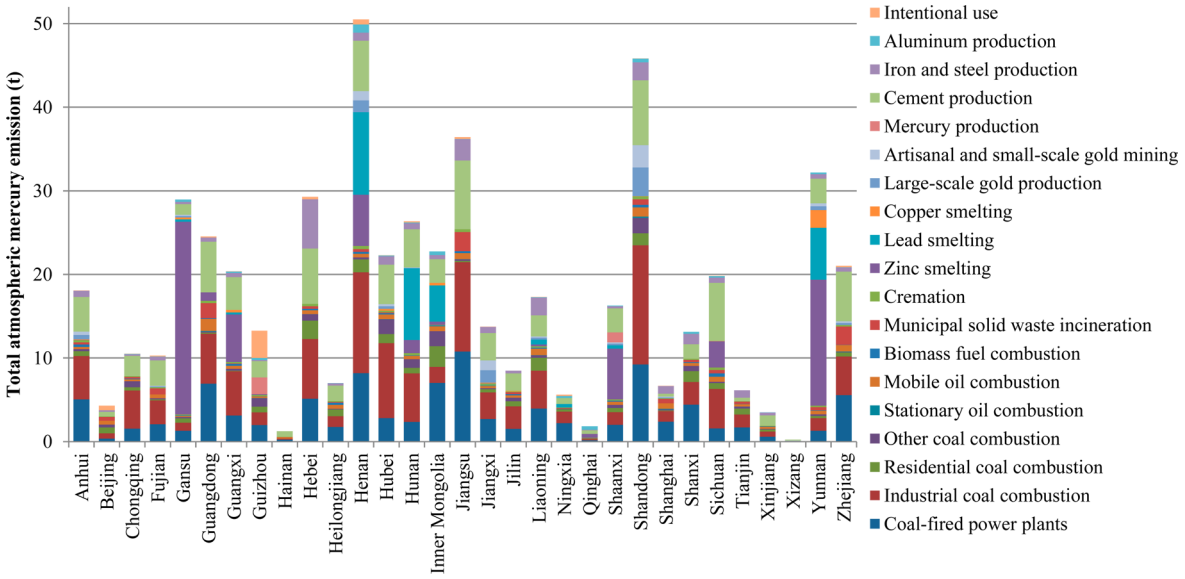

Figure 1. Provincial distribution of mercury emissions in China in 2010.

3. RESULTS AND DISCUSSION

3.1. Trend of Anthropogenic Atmospheric Mercury Emissions. Table 2 summarizes Hg emissions from different anthropogenic sources in China. The total atmospheric Hg emissions in China continuously increased from 356 t in 2000 to 538 t in 2010 with an annual average growth rate (AAGR) of 4.2%. During the same period, the activity levels of important sources such as coal-fired power plants, cement plants, and nonferrous metal smelters increased with AAGRs of more than 10%. This indicates that the air pollution control measures in key industrial sectors had a significant cobenefit on atmospheric Hg emission controls. The increasing rate of the total national Hg emission was generally consistent from year to year, but fluctuated more rapidly during 2002−2006 than during the other years. The highest increasing rate was triggered by the expedited development of coal-fired power plants, industrial coal combustion, zinc and lead smelting, cement production, and iron and steel production during 2002−2004, and the lowest increasing rate was a result of the widespread application of acid plants in nonferrous metal smelters during 2004−2006, which has a significant impact on Hg reduction. The emission intensities of coal combustion in both the power sector and the industrial sector decreased continuously from 2000 to 2010, down to 0.065 and 0.158 g Hg/t coal in 2010, respectively. However, the Hg emission intensity of cement production increased continuously from 0.027 to 0.052 g Hg/t cement produced within 2000−2010. Historical anthropogenic Hg emission trends from 2000 to 2010 are shown in Figure S2 in the Supporting Information, disaggregated by source type. In 2010, the largest contributor of Hg emission was the industrial coal combustion sector, accounting for 22.3% of the national total emissions. Coal-fired power plants, nonferrous metal smelting (zinc, lead, and copper), and cement production are responsible for 18.6%, 18.1%, and 18.3% of the total emissions, respectively. Iron and steel production, residential coal combustion, and mobile oil combustion contributed smaller but non-negligible portions of the total emissions. Although the contribution of waste incineration to total emission is small at present, its contribution could increase dramatically in the future due to its rapid growth rate. Artisanal and small-scale gold mining (ASGM) is illegal in China. Based on the estimation of China Gold Association, these illegal activities accounted for more than 10% of the total gold production back in 1990s but only 1−3% in 2010 owing to the explicit order of prohibition.[46,47] Therefore, only 6.8 t of Hg was emitted from this sector. However, it should be noted this estimate is with large uncertainties. Caustic soda production using a mercury cell electrolysis process has been controlled since 1996 and eliminated since the early 10th Five-Year Plan period (2001−2005).[48,49] Secondary emissions in the reutilization of by-products from coal combustion and unorganized emissions from nonferrous metal smelting are also potential emission contributors that are not considered in this inventory. Figure S3 in the Supporting Information shows the sectoral distribution of Hg emissions in China in 2010.

Coal-fired power plants, industrial coal combustion, nonferrous metal smelting, and cement production are crucial to the whole inventories. The total Hg emissions from coal-fired power plants increased significantly by 75% from 2000 to 2005, while remained about 100 t from 2005 to 2010 with a peak value in 2007 when electric power demand was increasing rapidly (shown in Figure S4 in the Supporting Information). This reveals the significant cobenefit mercury abatement of the sulfur dioxides (SO_2) control measures in the 11th Five-Year Plan for coal-fired power plants in China. The Hg emissions from industrial coal combustion were more or less 100 t from 2004 to 2007 and experienced a 20% jump in 2008 due to the sharp increase of the activity level. Mitigation of Hg emission occurred in this sector during the period of 2008−2010 due to PM and SO_2 control measures, although not as significant as in the power sector. More and more wet scrubbers were installed for simultaneous PM and SO_2 removal, and a small proportion of FF and WFGD was also employed. The Hg emissions from nonferrous metal smelting reached peak in 2003 and then decreased continuously. The Hg emission reduction in this sector was contributed by the adoption of acid plants in smelters since the acid producing process has high mercury removal efficiency. The application rates of acid plants for zinc, lead and copper smelters increased from 61%, 31%, and 61% in 2003 to 88%, 66%, and 96% in 2010, respectively. However, the Hg emissions from cement plants continuously grew because of

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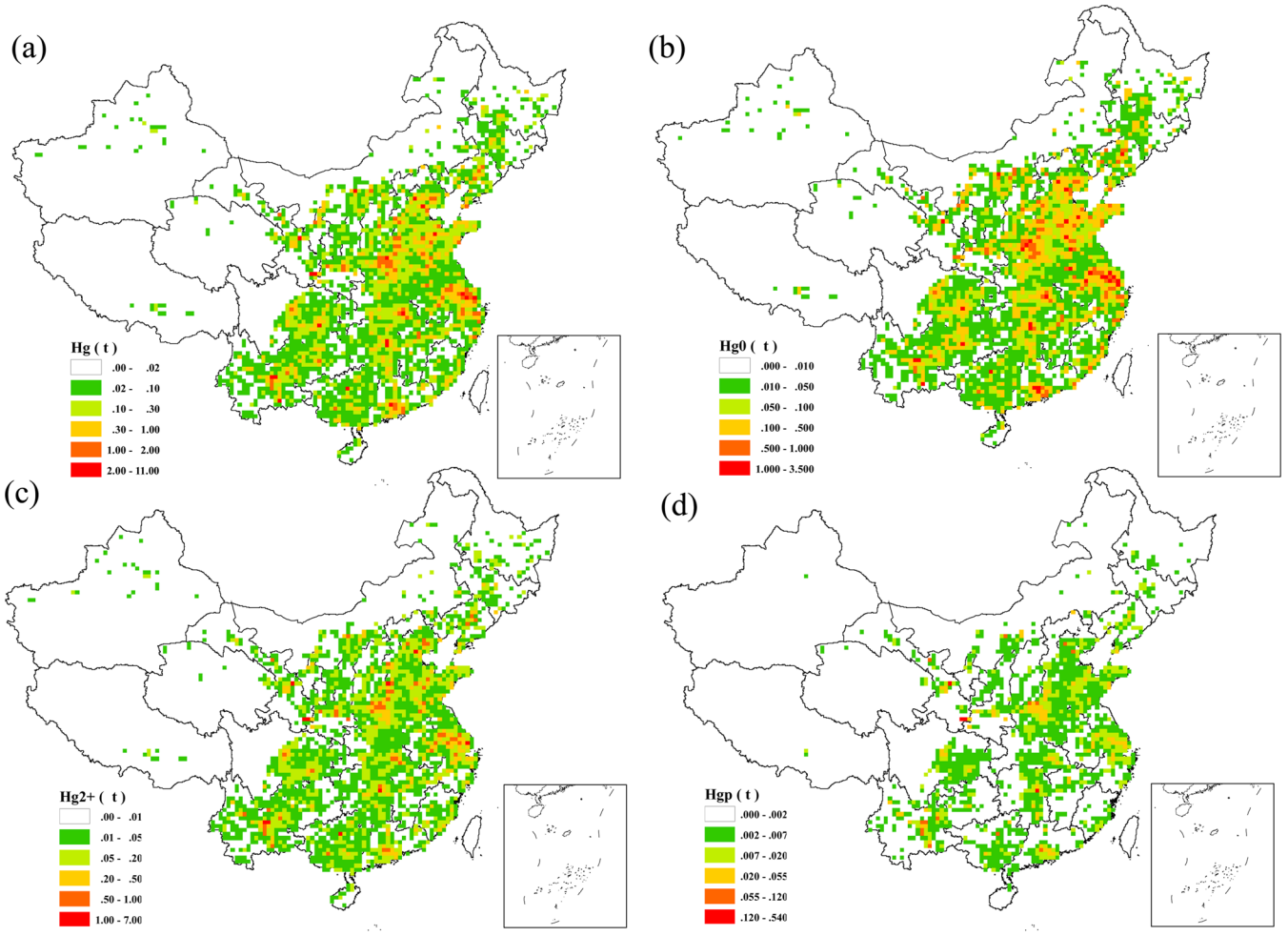

Figure 2. Gridded mercury emissions in China in 2010: (a) Hg_T; (b) Hg^0; (c) Hg^{II}; (d) Hg_p.

the increased cement production and the wide application of dry-process precalciner technology (which emits more Hg due to dust recycling[26]). The trends of the Hg emissions from the four sources are shown in Figure S5 in the Supporting Information.

3.2. Spatial and Species Distributions of Mercury Emissions in China. The provincial distributions of Hg emissions in China by sector in 2010 are shown in Figure 1. The top ten provinces are Henan, Shandong, Jiangsu, Yunnan, Hebei, Gansu, Hunan, Guangdong, Inner Mongolia, and Hubei, which accounted for almost 60% of the total Hg emissions in China. The North China Plain (NCP) region, including Hebei, Henan, Shandong, Jiangsu, and Anhui, is the heaviest mercury polluted area in China. The largest atmospheric Hg emitter, Henan Province, had a total emission of over 50 t, most of which was derived from industrial coal combustion, coal-fired power plants, lead and zinc smelting, and cement production. There are two key factors that will affect the range of environmental impacts of the emission sources: the height of the stack and the speciation of mercury in the exhausted flue gas. Hg emissions from "high-stack" sources, such as coal-fired power plants, industrial coal combustion, cement production, and iron and steel production, have a longer distance of atmospheric transport and thus result in larger range of environmental impact. In contrast, Hg emissions from "low-stack" sources, such as nonferrous metal smelting and intentional uses, have a shorter distance of atmospheric transport and intend for more local impacts. This has been proved in previous studies.[50] The provinces with large emission are divided into "high-stack-intensive" and "low-stack-intensive" ones. Hebei, Shandong, Jiangsu, and Guangdong are the "high-stack-intensive" type, whereas Yunnan, Gansu, and Hunan are the "low-stack-intensive" type. The mercury pollution in more developed areas, such as the North China region, the Yangtze River Delta (YRD) region, and the Pearl River Delta (PRD) region, had significant impacts on regional atmosphere. The mercury pollution in the southwestern, western and central parts of China does more harm to the local environment.

The speciation of mercury in exhausted flue gas has a more significant impact on the distance of Hg transport. Hg^0 has a lifetime of 0.5 to 2 years, whereas Hg^{II} and Hg_p have a lifetime of only hours to weeks.[2,51] Among the total Hg emissions in China in 2010, Hg^0, Hg^{II}, and Hg_p accounted for 58.1%, 39.4%, and 2.5%, respectively. Based on the locations of large point sources and the assignment of speciated Hg emissions, gridded emissions with a resolution of 36 × 36 km for large coal-fired power plants, nonferrous metal smelters, cement plants, and iron and steel plants regarded as point sources were obtained (see Figure S6 in the Supporting Information). The rest of the Hg emission sources were considered as nonpoint sources, and their speciated Hg emissions were also distributed to grids. The sum of point and nonpoint speciated emission are shown in Figure 2. The average national Hg^0, Hg^{II}, and Hg_p emission intensities from anthropogenic sources in 2010 are 19.1, 13.0,

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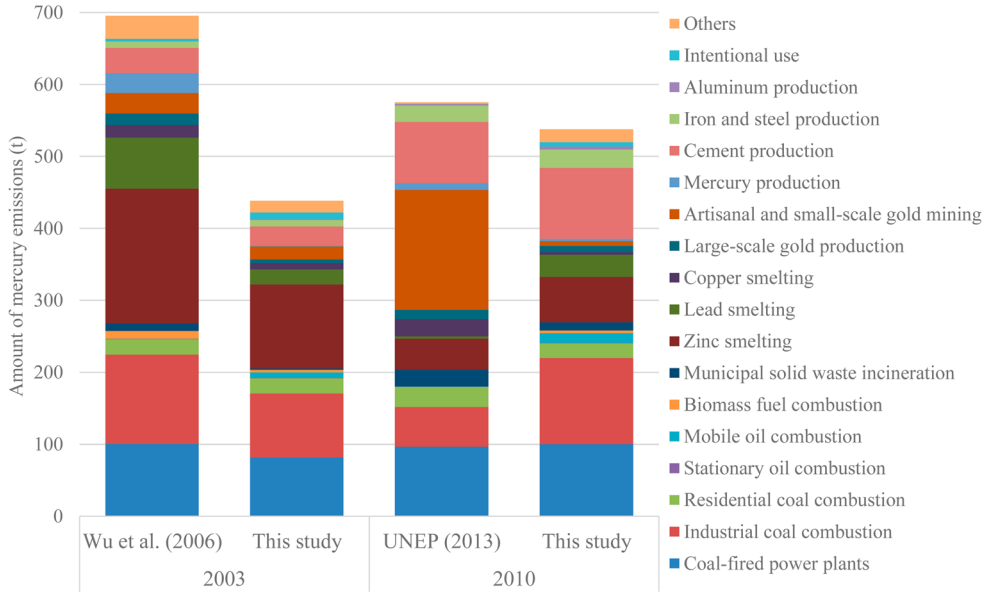

Figure 3. Comparison of Hg emission inventories between previous studies and this study for 2003 and 2010.

Table 3. Comparison of Mercury Speciation Profiles for Different Sectors in China in 2003 between This Study and the Previous Study

	Wu et al.[5] (2006)							this study						
	amount (t)				percentage (%)			amount (t)				percentage (%)		
	Hg^T	Hg^0	Hg^{II}	Hg_p	Hg^0	Hg^{II}	Hg_p	Hg^T	Hg^0	Hg^{II}	Hg_p	Hg^0	Hg^{II}	Hg_p
coal-fired power plants	100.1	20.0	78.1	2.0	20.0	78.0	2.0	81.4	52.4	27.6	1.3	64.4	34.0	1.6
industrial coal combustion	124.3	20.4	88.5	15.4	16.4	71.2	12.4	89.0	54.0	29.6	5.3	60.7	33.3	6.0
residential coal combustion	21.7	2.0	0.7	19.1	9.0	3.0	88.0	21.1	17.6	1.5	2.1	83.1	6.9	10.0
biomass fuel combustion	10.7	10.3	0.0	0.4	96.0	0.0	4.0	3.6	2.7	0.2	0.8	74.4	4.8	20.8
municipal solid waste incineration	10.4	9.9	0.0	0.4	96.0	0.0	4.0	1.8	1.8	0.0	0.1	96.0	0.0	4.0
zinc smelting	187.6	150.1	28.1	9.4	80.0	15.0	5.0	116.6	52.9	57.9	5.8	45.3	49.7	5.0
lead smelting	70.6	56.5	10.6	3.5	80.0	15.0	5.0	21.2	14.7	5.4	1.1	69.4	25.6	5.0
copper smelting	17.6	14.1	2.6	0.9	80.0	15.0	5.0	8.6	5.1	3.1	0.4	59.0	36.0	5.0
large-scale gold production	16.2	12.9	2.4	0.8	80.0	15.0	5.0	5.0	4.0	0.7	0.2	80.0	15.0	5.0
mercury production	27.5	22.0	4.1	1.4	80.0	15.0	5.0	1.2	1.0	0.2	0.1	80.0	15.0	5.0
cement production	35.0	28.0	5.3	1.8	80.0	15.0	5.0	26.7	18.1	7.6	1.1	67.6	28.4	4.0
iron and steel production	8.9	7.1	1.3	0.4	80.0	15.0	5.0	8.9	2.9	5.6	0.4	32.1	62.9	5.0

and 0.8 $\mu g/m^2$. The YRD region suffered from extremely high emission intensities for all the Hg species due to its dense industries and small area, 181, 108, and 5.1 $\mu g/m^2$ for Hg^0, Hg^{II}, and Hg_p, respectively. Heavy Hg^0 emissions covered the North China region (98 $\mu g/m^2$) and the PRD region (81 $\mu g/m^2$). The emission of Hg^{II} is even more widely distributed than that of Hg^0, with substantial amount in North China region (60 $\mu g/m^2$) and the Southwest China region (50 $\mu g/m^2$) caused by large nonferrous metal smelters and cement plants. Regional Hg_p emissions also occurred in the North China region (3.6 $\mu g/m^2$) and the Southwest China region (3.4 $\mu g/m^2$).

3.3. Comparisons with Existing Hg Emission Inventories for China. Compared the results from this study with the existing literature[5,8] for the same inventory year, both the sectoral and species distributions are quite different between this study and previous ones. The sectoral distributions were compared between this study and Wu et al.[5] (2006) for the 2003 inventory and between this study and AMAP and UNEP[8] (2013) for the 2010 inventory, as shown in Figure 3. The most significant difference in the 2003 inventory occurred in nonferrous metal smelting (Zn, Pb, and Cu). The total Hg emissions from this sector was estimated to be 146 t in 2003 in this study, 47% lower than the estimate from Wu et al. (2006). The estimation for this sector was based on 8 on-site measurements of Chinese nonferrous metal smelters, and the high Hg removal efficiency in APCDs in smelters were also confirmed by other recent studies.[7,11,12] The estimate of Hg emissions from coal combustion in this study was 22% lower than that from Wu et al. (2006), which was mainly due to the difference on Hg removal efficiency of wet scrubber for PM control. The Hg emission from MSW incineration was estimated to be 11.6 t in 2010 in this study, almost half of that in the UNEP report[8] but within the variation range (4.7–36.7 t) of the recent studies,[15−17] which indicates large uncertainties in Hg emission from this sector. The two biggest differences in the 2010 inventory between this study and the UNEP report were artisanal and small-scale gold mining (ASGM) and industrial coal combustion. There is a good chance that the UNEP report on the China inventory has a significant overestimation of the ASGM sector and an underestimation in the sector of industrial coal combustion. Because official data from China on the activity levels of these

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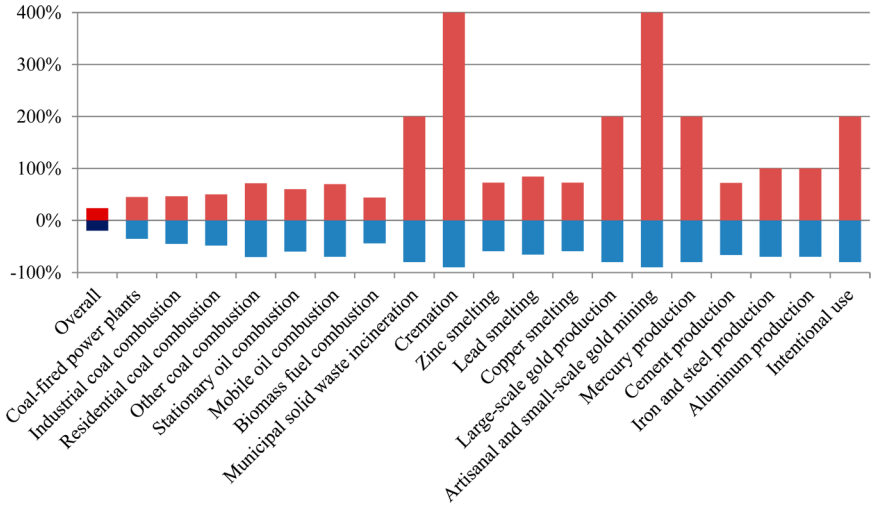

Figure 4. Uncertainty ranges of mercury emissions from different sources in China.

two sectors were not available in the UNEP global mercury assessment, the estimation was mostly based on the presumption from other countries, whose circumstances are quite different from China.

Table 3 shows the comparison of the profiles of speciated mercury from different sectors in China in 2003 between this study and the previous study.[5] In the previous study, Hg^{II} was the dominant Hg species for coal-fired power plants and industrial coal combustion, accounting for over 70% of the total Hg emissions, and Hg_p took the lead for the sector of residential coal combustion (88%). With the updated Hg speciation data in this study, Hg^0 turned out to be the dominant form of Hg emission for coal combustion, contributing to 64%, 61%, and 83% of the total Hg emissions from power, industrial, and residential coal use, respectively. The Hg^0 proportion of the exhausted flue gas from biomass fuel combustion was 74% in this study, lower than the estimation of the previous study (96%). The total emissions from nonferrous metal smelting and large-scale gold production are 38%−70% lower in this study than those in previous studies, which was caused by the high mercury removal efficiency of acid plants in smelters (86%−99%). The proportion of Hg^{II} in the exhausted flue gas from zinc smelting is 50% in this study, much higher than the estimation of the previous study (15%). The same cases also occurred to cement production and iron and steel production that 28% and 63% of the total emissions from these two sectors are Hg^{II} in this study. There are no updated profiles for the sectors of large-scale gold production and mercury production. However, the total amount of Hg emissions from these three sectors changed significantly due to the updated Hg emission factors. The significant change of Hg speciation in dominant emission sources will drastically change the estimates of long-range transport of the Hg pollution from China and the contribution of Hg emissions in China to the global background.

3.4. Uncertainties in the Mercury Emission Inventory. Using the approach developed in this study for uncertainty analysis, uncertainties of Hg emission estimates for coal combustion, nonferrous metal smelting, and cement production were determined. Based on the probabilistic technology-based emission factor model and the detailed data for parameters in the Hg emission factors, the uncertainty ranges for dominant sources were largely reduced, including coal-fired power plants (−35%, +45%), industrial coal combustion (−45%, +47%), residential coal combustion (−48%, +50%), zinc smelting (−59%, +72%), lead smelting (−65%, +84%), copper smelting (−59%, +72%), and cement production (−66%, +72%). The uncertainty ranges for sources in Tier 1 and Tier 2 were inherited from previous studies.[4,18] The overall uncertainty in the new emission inventory for China was estimated to range from −20% to 23% which was significantly reduced compared with those from previous studies[4,5] (±44%). The uncertainty ranges for all the sectors are shown in Figure 4. Coal combustion, nonferrous metal smelting, and cement production constitute over 80% of the total Hg emission inventory. The significant decrease of uncertainty level in this study is mainly due to the significant decrease of uncertainty levels of these dominant sources, especially nonferrous metal smelting and cement production. Previous studies[4,5] adopted single emission factors for nonferrous metal smelting and cement production from international experience, while this study, for the first time, used the results from on-site measurements conducted in China and adopted the technology-based emission factor model, which improved the accuracy of the inventory.

The uncertainties in the estimation of coal mercury contents from the two largest coal-producing provinces, Shanxi and Inner Mongolia, contributed 31% and 24% to the uncertainties of Hg emissions from coal-fired power plants, respectively, and contributed 40% and 42%, respectively, to the uncertainties of Hg emissions from residential coal combustion. The uncertainty of Hg content in coal from Shanxi province also accounted for 36% of the uncertainties of Hg emissions from industrial coal combustion, while another 33% came from the uncertainties in the mercury removal efficiency of wet scrubber (WS). For nonferrous metal smelting, most of the uncertainties originated from mercury concentration in metal concentrates from large concentrate-producing provinces, Gansu, Shaanxi, and Yunnan contributing 33%, 20%, and 18%, respectively, for zinc smelting. Uncertainties in the estimation of mercury concentration in limestone were key factors contributing to the uncertainties of Hg emission from cement production by 97%.

With the adoption of the novel methodology and the reduction of uncertainty levels of key parameters for the estimation of dominant Hg emission sources including coal combustion, nonferrous metal smelting and cement production, the accuracy of Hg emission inventories for China was

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significantly improved in this study. Uncertainties of Hg concentrations in coal, metal concentrates and limestone are major contributors to the uncertainties of Hg emission estimates of coal combustion, nonferrous metal smelting and cement production, respectively, and consequently accounted for over 60% of the overall uncertainties of the Hg emission inventories of China. Based on current knowledge or practices, further reduction of uncertainties from Hg contents in fuel or raw material is not practical. The uncertainties of activity levels and the APCD application rates contributed more or less 20% to the overall uncertainties. The rest uncertainties were caused by those from the mercury removal efficiencies of APCD combinations, which is possible to be further reduced. The most probable APCD combinations for China in the future are SCR+ESP/FF+WFGD for coal-fired power plants, FF+WFGD for coal-fired industrial boilers, acid plant with the DCDA process for nonferrous metal smelters and dry-process precalciner technology with dust recycling for cement plants, respectively. More research on the mechanisms of Hg transformation in flue gas across these APCDs needs to be conducted to further reduce the uncertainties for cement production. MSW incineration, a potential dominant Hg emission source in the future in China, has a large uncertainty level (−80%, +200%). There is an urgent need for on-site measurements for the MSW incinerators in China. Extensive and dedicated field work is required for potential dominant Hg emission sources in China in the future.

■ ASSOCIATED CONTENT

Supporting Information

Previous inventories for the atmospheric mercury emissions in China, methods for the four tiers of emission sources, mercury emission factors for the mercury emission sources in Tier 2, mercury concentration in fuel/raw material, mercury speciation in exhausted flue gas, activity levels and APCD application rates of mercury emission sources in China, methodology for uncertainty analysis, anthropogenic mercury emissions in China from 2000 to 2010, and gridded mercury emissions from different sources in China in 2010. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Tel.: +86 10 62771466. Fax: +86 10 62773597. E-mail: shxwang@tsinghua.edu.cn (S. Wang).

Author Contributions

[§]Joint first authors. L.Z. and S.W. contributed equally to this work.

Notes

The authors declare no competing financial interest.

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