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A Highly Resolved Mercury Emission Inventory of Chinese Coal-Fired Power Plants

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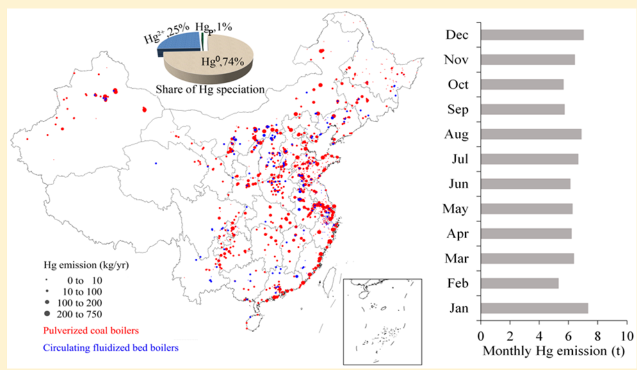
Supporting Information

ABSTRACT: As the largest coal consumer in China, the coal-fired power plants have come under increasing public concern in regard to atmospheric mercury pollution. This study developed an up-to-date and high-resolution mercury emission inventory of Chinese coal-fired power plants using a unit-based method that combined data from individual power plants, provincial coal characteristics, and industry removal efficiencies. National mercury emissions in 2015 were estimated at 73 tons, including 54 tons of elemental mercury, 18 tons of gaseous oxidized mercury and 1 ton of particle-bound mercury. Pulverized coal boilers emitted 65 tons, mainly in the coastal provinces and coal-electricity bases. Circulating fluidized bed boilers emitted 8 tons, mainly in Inner Mongolia and Shanxi Province. The average mercury emission intensity over the Chinese mainland was 18.3 g/GWh, which was similar to the limit for low-rank coal-fired units in the United States. The overall uncertainty of national mercury emission was estimated to be −19% to 20%, with the mercury content in coal being the major contributor. In most provinces, monthly mercury emissions generally peaked in December and August. However, monthly partition coefficients of southwest China were obviously lower than other regions from June to October due to the high proportion of hydropower generation.



1. INTRODUCTION

Mercury has aroused global concern because of its toxicity, persistence, long-range transport, and bioaccumulation in the environment, which make mercury a potential threat to both humans and ecosystems.[1,2] The global pollution of mercury led to the signature of the Minamata Convention on Mercury in October 2013, which comes into force in August 2017.[3,4]

As the second largest source of anthropogenic mercury emission to the air, power plants were estimated to emit 16% of the global mercury emission in 2010.[5] In China, mercury emissions from coal-fired power plants have been estimated in many national, regional, and global inventories. Wu et al. (2006)[6] estimated that the national Mercury emissions from coal-fired power plants grew at an annual growth rate of 6% during 1995−2003, and by 2003, it amounted to 100 tons.[6] Zhang et al. (2015)[7] yielded an historical inventory of coal-fired power plants from 2000 to 2010 with a growth rate of 6%, the peak (105 tons) of which occurred in 2007.[7] Wu et al. (2016)[8] found the mercury emission of coal-fired power plants to be 82 tons in 2014.[8]

After the implementation of the "Air Pollution Prevention and Control Action Plan (2013)" and the "Ultra-Low Emission and Energy Saving of Coal-fired Power Plant Plan (2015)", selective catalytic reduction (SCR), wet electrostatic precipitators (WESPs) and advanced electrostatic fabric filters (ESP-FFs) have become popular.[9,10] Normally, mercury in flue gas exists in three operationally defined forms, namely, elemental mercury (Hg^0), gaseous oxidized mercury (Hg^{2+}), and particle-bound mercury (Hg_P).[11] SCR catalysts can promote the oxidation of some of the Hg^0 to Hg^{2+}, which contributes to additional reduction of Hg^{2+} in FGD gypsum. Hg_P absorbed in fine particulate matter can be effectively captured by ESP-FF,

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which is highly cobeneficial for mercury removal in coal-fired power plants.

This study develops a high-resolution mercury emission inventory for Chinese coal-fired power plants and explores its temporal and spatial characteristics using a unit-based method. In addition, the mercury emission intensity by province is calculated and compared with the emission limits of low-rank/high-rank coal-fired units in the U.S. to analyze the potential mitigation. The method allows power plants to act as point-sources for atmospheric models. In addition, the results can help governments establish criteria to identify "relevant sources", as required in the Minamata Convention on Mercury.

2. MATERIALS AND METHODS

Most of the previous mercury emission inventories were compiled by a province-based, bottom-up method.[6,8,12−19] Zhao et al. (2015)[19] evaluated the effects of China's pollution controls on interannual trends of atmospheric mercury emissions from 2005 to 2012 with a provincial bottom-up method.[19] Wu et al. (2016)[8] used a technology-based approach to compile a consistent series of China's atmospheric mercury emissions at the provincial level from 1978 to 2014.[8] Streets et al. (2005)[13] evaluated the mercury emissions from 283 power plants based on field testing results of the U.S. Environmental Protection Agency (EPA).[14] Zhang et al. (2015)[7] developed the emission inventory of coal-fired power plants in 2010 based on domestic field testing and the provincial constitution of APCDs.[7] Mercury emissions from Chinese coal-fired power plants in major global-scale inventories, such as those by UNEP/AMAP and EDGAR, were based on the results in Zhao et al. (2016) and Streets et al. (2005).[5,20] However, both studies ignored that the cobenefit removal efficiency of APCDs significantly varies among different coal types and that the type of APCDs applied in pulverized coal (PC) boilers and circulating fluidized bed (CFB) boilers are different.[21,22] Furthermore, APCDs types have changed, most Chinese power plants have installed selective catalyst reduction (SCR), flue gas desulfurization (FGD), and high efficiency dust collects to meet the ultralow emission standard, which results in higher mercury removal efficiency. Therefore, previous studies fail to represent the current tempo-spatial characteristics of mercury emissions from coal-fired power plants.

In contrast to previous studies, this study took the impacts of feed coal types (anthracite, bituminous coal, lignite coal and coal gangue) on mercury emission into calculation. We compiled the mercury emission inventories for PC boilers and CFB boilers respectively to ensure responsive policy-making. The mercury removal efficiencies of both APCDs for ultralow emission − SCR+ESP-FF+WFGD and SCR+ESP+WFGD+WESP were updated. A unit-based database for the year of 2015 was established, including coal consumption, Hg content in the feed coal, boiler type, coal type, APCD type, and geographical location of 1472 PC boilers and 345 CFB boilers.[23−26] In addition, this was the first study to develop a monthly mercury emission inventory from coal-fired power plants at the province level and explore the diurnal and hourly variation. Such methodology could be applied to other mercury emission sources, such as cement clinker production, non-ferrous metal smelters and so on, which would provide emission input with high tempo-spatial resolution for mercury chemical transport models (CTMs).

We updated the unit-based method, which is shown as follows.[7,14,20]

$$E_{i,j,t} = A_{i,j} \times M_i \times (1 - Q_i \times w) \times R \times (1 - \eta_{i,j}) \times T_{i,t} \quad (1)$$

$$E_{i,t} = \sum_j E_{i,j,t} \quad (2)$$

$$E_{\text{total}} = \sum_i \sum_t E_{i,t} \quad (3)$$

where $E_{i,j,t}$ is the mercury emission of power plant j located in province i for the month t (t); $A_{i,j}$ is the annual coal consumption of power plant j (Mt); M is the mercury content of feed coal (g/t); Q is the percentage of washed coal; w represents the mercury removal efficiency of coal washing; R is the release ratio; η represents the cobenefit removal efficiency of APCD equipped by power plant j, which is affected by feed coal type and boiler type; T is the monthly partition coefficient; Q, w, R, η, and T are entered as proportions (%/100%).

2.1. Mercury Content in Coal. As shown in Supporting Information (SI) Table S1, the mercury content in raw coals from different provinces varied dramatically owing to the different geological coal-forming environments and coal-forming plants.[27,28] In addition, coal reserves were unevenly distributed in China. As a result of the uneven geographic distribution between coal production and consumption, the coal consumed in developed provinces is transported from nearby coal-producing provinces or even imported from other countries. In 2015, China imported 193 Mt of coal (accounting for ∼3% of the total amount of coal in China) from Indonesia (74 Mt), Australia (71 Mt), North Korea (19 Mt), Russia (15 Mt), and Mongolia (14 Mt).[24,29] The mercury content in thermal coal imported from Indonesia and Australia is 0.05 g/t and 0.02 g/t, which is lower than that in Chinese coal.[30] Due to the low mercury content in the coal, the corresponding mercury emission was less.

To obtain the Hg content in coal burned in each province, we compiled a matrix including 30 provinces and other coal-exporting countries based on official statistical data from the China Energy Statistical Yearbook (2016) (see more details in SI Table S2).[25] As shown in Figure 1, coal consumed in Shandong, Hebei, Jiangsu, and Tianjin was mainly transported from Inner Mongolia; part of the coal consumed in Henan, Anhui and Guangdong was from Shanxi; part of the coal consumed Hubei, Hunan, and Fujian was transported from Shaanxi; and about half of the imported coal was combusted in Guangdong and Fujian. The provincial weighted-average Hg content in the consumed coal, shown in SI Table S1, was determined by combining this information with the database for the mercury content in raw coal compiled by Zhang et al. (2012).[15,25] The lowest Hg content in the feed coal was obtained in Xinjiang (0.05 g/t), and the highest in Chongqing (0.37 g/t).

In China, coal gangue is the industrial residue of coal mining and washing. To effectively utilize the calorific value and alleviate the problem of land occupation, coal gangue is extensively combusted in power plants as a raw material. This is the first study that developed a mercury emission inventory from coal gangue. Zhai et al. (2015)[31] collected coal gangue samples from four coal mines in Shanxi and found that sulfide-bound Hg was the dominant form in coal gangue and the Hg content was higher than that in raw coal.[31] Wang et al. (2016)[32]

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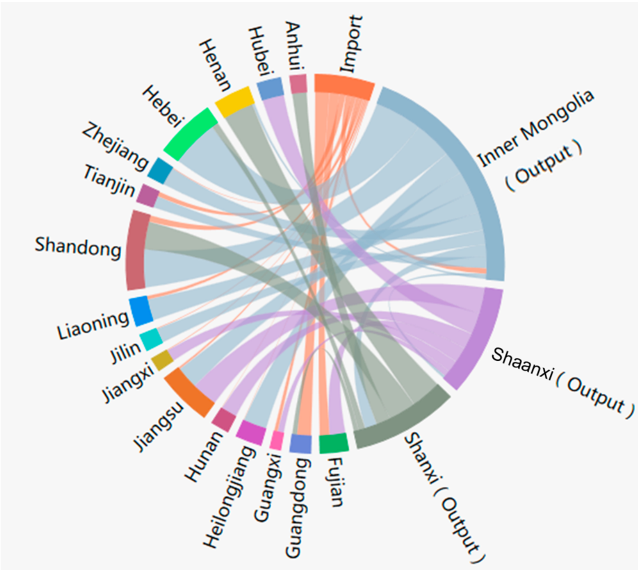

Figure 1. Main transport flow of raw coal in 2015.

collected and analyzed the Hg content in representative coal gangue samples from large coal mines in Shaanxi, Shanxi, and Shandong. The Hg content in the coal gangue samples was found to range from 0.14 to 0.34 g/t, which is slightly higher than that in raw coal.[32] The median (0.24 g/t) was applied to the calculation in this study.

2.2. Mercury Removal Efficiencies and Speciation Profiles. The Hg removal rates of existing conventional APCDs vary significantly depending on the coal type, boiler type and APCD type and range from 19% to 90%, as shown in SI Table S3. Sixty-nine on-site tests of the cobenefit mercury removal efficiencies from existing studies are summarized in Table 1 (more details can be seen in SI Table S4).[7,8,21,32−39] The uncertainty bounds of the APCD removal efficiencies for the uncertainty analysis are summarized in SI Table S4. Generally, PC boilers burning anthracite demonstrated higher Hg removal than similarly equipped boilers burning bituminous coal, which could achieve higher mercury removal than those burning lignite coal.[15,40] CFB boilers equipped with ESP or FF showed a higher average cobenefit removal efficiency than PC boilers also equipped with ESP or FF.[7,19]

Nearly all of the Hg_P could be simultaneously captured by ESP/FF, which led to an approximately 18−30% reduction of flue-gas mercury.[21] When the flue gas was dragged through wet flue-gas desulfurization (WFGD), 67−98% of the Hg^{2+} could be absorbed in the scrubber solution and then retained in the gypsum.[21,41] SCR catalysts could promote the oxidation of part of the Hg^0 to Hg^{2+} and thus alter the speciation of Hg in flue gas, especially for coal with a high Cl content.[15] The share of Hg^{2+} in flue gas could increase by more than 10% across the SCR bed, which contributed to the further reduction of the total mercury in FGD gypsum. Therefore, the APCD type greatly effects the mercury removal efficiency and Hg speciation profiles in the flue gas. SI Figure S1 shows the mercury mass transfer flow from the feed coal to plant emission.

Hg^0 can be retained for several months with a long transport distance until settling out through dry and wet deposition processes, while Hg^{2+} and Hg_P can be retained for only hours to weeks.[42,43] To identify the current status and future fate of Hg from coal-fired power plants, Table 1 summarizes the 30 existing Hg speciation profiles of coal-fired flue gas. Details are given in SI Table S5.[7,8,21,32−38]

Coal washing primarily aims to minimize the ash or sulfur content in raw coal but can also decrease the Hg content with a cobenefit removal efficiency of 0−60%.[6,44] In Guizhou and Shaanxi, the proportions of washed coal in coal-fired power plants were up to 37% and 17%, respectively.[25] This article assumes a Hg removal efficiency of 30% in the calculation.

2.3. Consumption of Different Types of Coal. In China, coal burned for the generation of electricity is mainly anthracite, bituminous coal, lignite coal, and coal gangue, 66% of thermal coal is bituminous coal, mainly produced from Inner Mongolia, Xinjiang, and Shaanxi; 19% is anthracite, mainly from Guizhou and Shanxi; 13% is lignite coal, mainly from the mining area of northeast Inner Mongolia; and 3% is coal gangue, mainly from Inner Mongolia and Shanxi.[26,45−49] In 2015, the national amount of coal gangue combusted for electric generation reached 45.72 Mt. In Shanxi and Inner Mongolia, the amount of coal gangue in the feed coal of power plants reached 10.44 Mt and 7.53 Mt, respectively.[25] Combined with the coal flow matrix, the composition of the feed coal type by province is

Table 1. Removal Efficiencies and Speciation Profiles of Mercury in Coal-Fired Power Plants

APCD		removal efficiencies, η (%)				speciation profiles (%)		
category	release ratio, R (%)[a]	anthracite	bituminous	lignite	gangue	Hg^0	Hg^{2+}	Hg_P
(PC) ESP+WFGD	99	81.4	63.4	46.2		81.9	17.7	0.4
(PC) FF+WFGD			84.2			81.1	17.5	1.4
(PC) ESP-FF+WFGD			87.2			87.3	12.0	0.7
(PC) SCR+ESP+WFGD		80.0	70.4	56.6		77.9	21.9	0.2
(PC) SCR+FF+WFGD			87.8			34.5	62.2	3.3
(PC) SCR+ESP+WFGD+WESP			95.1			65.8	32.8	1.4
(PC) SCR+ESP-FF+WFGD		98.1	97.6	92.3		74.3	22.8	2.9
(PC) SCR+ESP+SW-FGD[b]			74.1			77.9	21.8	0.2
(PC) NID+ESP[b]			88.5			0.1	81.0	17.9
(PC) ESP+CFB-FGD+FF			66.0			66.7	33.2	0.1
(CFB) ESP	99		73.1	56.0	72.9	71.9	27.5	0.6
(CFB) FF			92.5	59.0	76.0	82.0	17.5	0.5
(CFB) SNCR+ESP+WFGD[c]			98.1			51.2	47.9	0.9

[a]The release ratio is compared with the total amount of corresponding mercury in the feed coal. [b]Sea water flue-gas desulfurization (SW-FGD); novel integrated desulphurization (NID); selective non-catalytic reduction (SNCR).

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given in Figure 2. As the top coal consumer for electric generation, Inner Mongolia's feed coal was composed of 48%

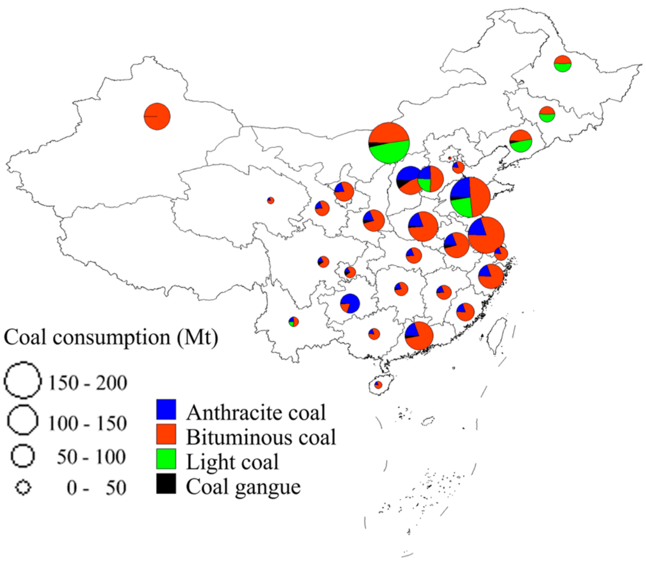

Figure 2. Composition of different types of coal in Chinese power plants, 2015.

lignite coal, 48% bituminous coal, and 4% coal gangue. Due to the input of lignite coal from Inner Mongolia and anthracite from Shanxi, Shandong's feed coal was composed of 24% anthracite, 49% bituminous coal, 24% lignite coal, and 3% coal gangue. For the main anthracite producers, Shanxi and Guizhou, the proportion of anthracite in the coal was 45% and 79%.

2.4. Distribution of Boiler Types and APCDs. In China, PC and CFB boilers are the major boiler types. By the end of 2015, PC boilers comprised 90% of the installed capacity in coal-fired power plants.[23,25] Since the 1980s, CFB boilers have been rapidly developed in China as a key clean coal technology for controlling SO_2 and NO_x pollution.[50] Moreover, they can be used for the comprehensive utilization of coal gangue and slime (a byproduct in the process of coking coal production and one type of inferior coal). The total capacity of CFB boilers has grown to approximately 110 GWe, and a CFB unit with the largest installed capacity of 600 MW has begun trial operation in Sichuan.[24] In-furnace desulphurization by spraying calcium and a flue-gas electrostatic precipitator is generally applied in CFB boilers to control SO_2 and particulate matter (PM) pollution.[41,51,52]

Different types of APCDs show different performances in the add-on synergistic removal of mercury from coal-fired flue gas.[15,32] ESP or FF is applied as a dust collector, and FF achieves more stable and higher performance regardless of coal type, resulting in greater mercury emission reduction. To avoid the disadvantageous high pressure loss, hybrid dust collectors (combining ESP and FF, ESP-FF) have been increasingly applied.[53] WESP represents another advanced dust collector technology, which is equipped after the wet desulphurization stage and can collect fine particles and mists, as well as hazardous trace elements.[54,55] In response to the stringent PM emission standard (GB13223−2011), ESP-FFs and WESPs are being increasingly retrofitted or built in coal-fired power plants.[56]

WFGD and SCR are commonly utilized in pulverized coal boilers, where both comprise over 95% of the national installed capacity of PC boilers. Currently, the proportion of both APCDs for ultralow emission − SCR+ESP-FF+WFGD and SCR+ESP+WFGD+WESP − increased to 18% and 4% in Chinese coal-fired power plants in 2015.[23] The types of APCDs by province are given in Figure 3.

3. RESULTS AND DISCUSSION

3.1. Spatial Characteristics of Mercury emissions. By the end of 2015, the national installed capacity of coal-fired power plants had increased to 899 GW, generating 3898 TWh of electricity. The corresponding coal consumption in the Chinese mainland was 1792 Mt. Atmospheric Mercury emissions from coal-fired power plants in 2015 were estimated to be 73 tons (−19%, + 20%), including 54 tons of Hg^0, 18 tons of Hg^{2+}, and 1 ton of Hg_P. As high-stack sources of mercury emission, coal-fired power plants significantly impact the regional atmosphere.[57] Inter-regional mechanisms for the joint prevention and control of air pollution have been implemented since 2010. Coal-fired power plants bore the brunt of the mitigation of air pollutants, resulting in decommissioning a number of small and outdated coal-fired

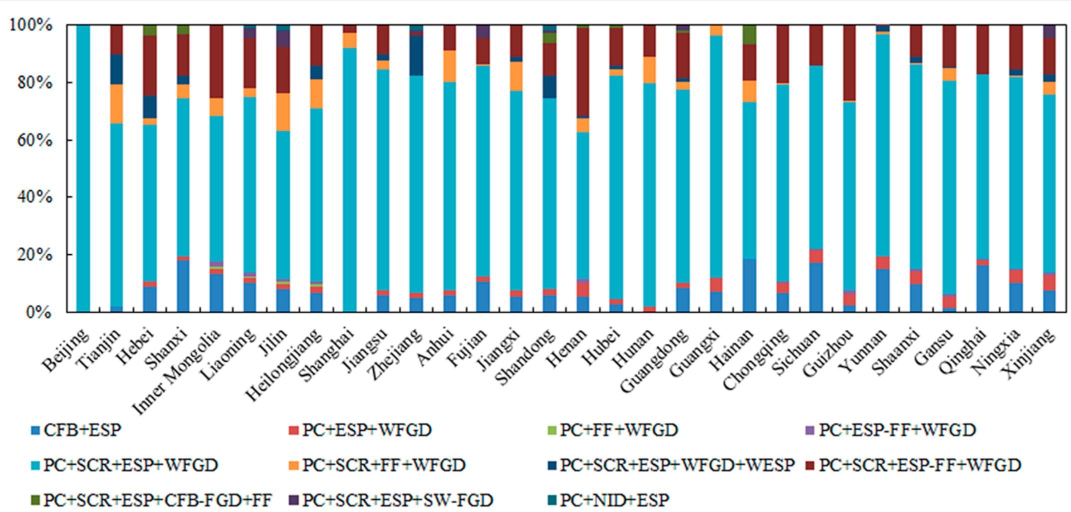

Figure 3. Types of APCDs by province in coal-fired power plants, 2015.

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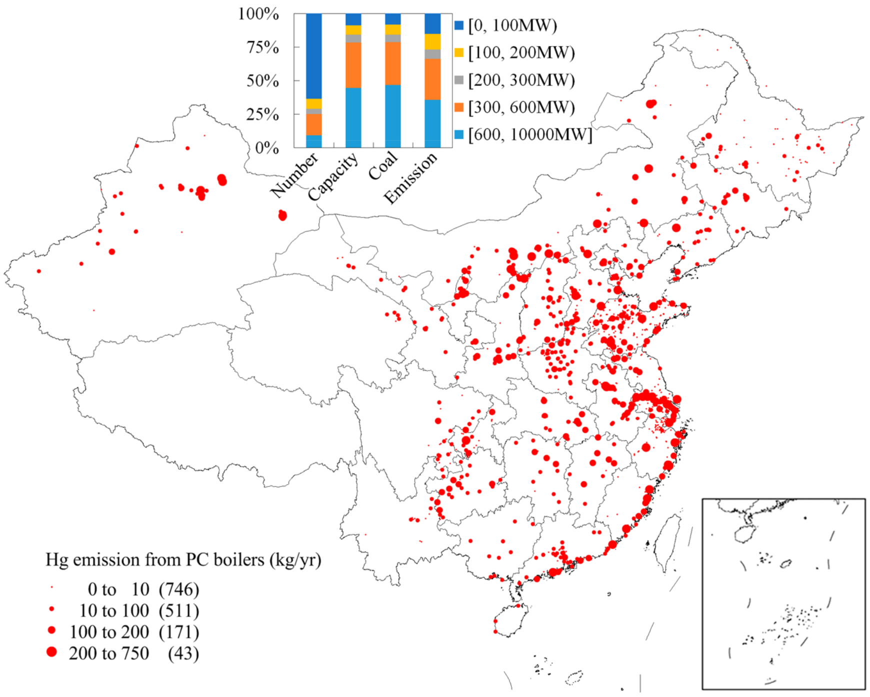

Figure 4. Geographical distribution of Mercury emissions from PC boilers (the size of the circle represents the annual mercury emission and the numbers inside the parentheses are the quantities).

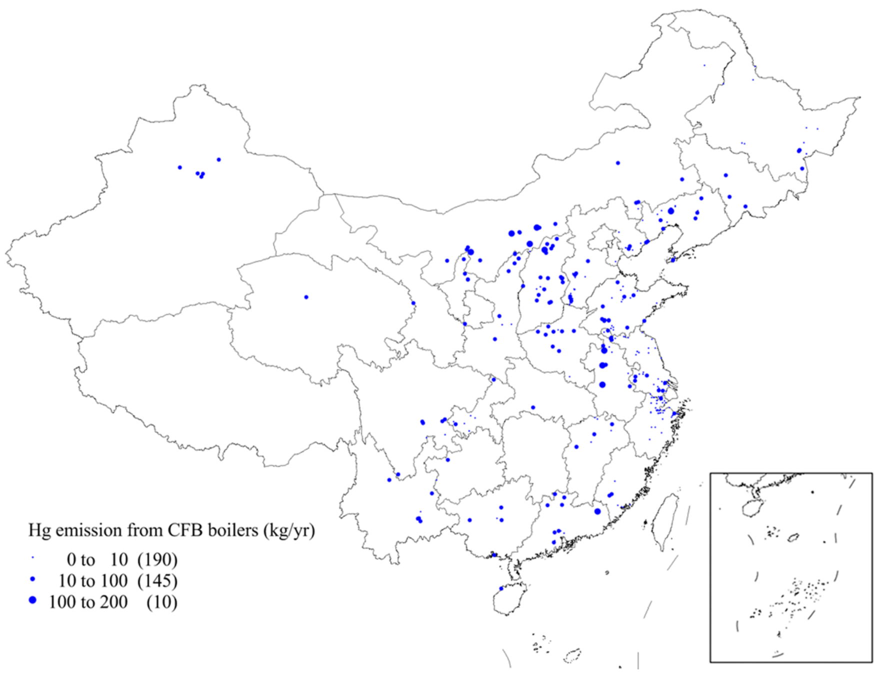

Figure 5. Geographical distribution of Mercury emissions from CFB boilers (the size of the circle represents the annual mercury emission and the numbers inside the parentheses are the quantities).

power plants in developed regions, such as the Beijing-Tianjin-Hebei region, the Yangtze River Delta region, and the Pearl River Delta region.[58]

Figures 4 and 5 illustrate the spatial distribution of mercury emissions from 1472 PC boilers and 345 CFB boilers, respectively. Atmospheric mercury emissions from PC boilers in 2015 were estimated to be 65 tons (−19%, + 21%), accounting for 89% of the total amount. Most PC boilers were concentrated in or around the coastal provinces, which have high urban and industrial power demands, such as Shandong, Jiangsu, and Zhejiang Province. In the implementation of a program to "Transfer Electricity from West to East", a series of coal-electricity bases were built in Inner Mongolia, Shanxi, and Shaanxi, which are rich in coal resources. As the main power sources in the program, numerous large coal-burning plants were built in these coal-electricity bases. In 2015, 43 PC boilers emitted more than 200 kg/yr of mercury, and the largest emitter, which emits 741 kg/yr, was located in Inner Mongolia.

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According to the statistics presented in the histogram in Figure 4, generator units with sizes ≥600 MW had a capacity of 422 GW, accounting for 44% of the total installed capacity. Mercury emissions from these units were estimated to be 24 tons, accounting for 36%. However, 23 tons of Hg were emitted from generator units with sizes ≤300 MW, and the combined capacity was only 204 GW. Notably, 4003 small generator units with sizes ≤100 MW were still in operation in 2015, accounting for 63% of the total. Thus, there is still room to expand the "Substitution of Smaller Units with Big Ones" policy, which is conducive to mercury reduction. The composition of the installed capacities of different-sized units by province is shown in SI Figure S2.

CFB boilers combusted 163 Mt of feed coal and emitted 8 tons of atmospheric mercury in 2015, accounting for 11% of the total emission. Furthermore, 10 Mt of coal gangue was produced in Shanxi from coal mining and washing. Due to their effective utilization of low-grade coals, a few CFB boilers were established in coal-producing regions, such as Inner Mongolia and Shanxi, as shown in Figure 5. These CFB boilers combusted 1797 tons of gangue, accounting for 39% of the national amount of gangue burned in power plants. The largest emitter, which emitted 149 kg, was also located in Inner Mongolia. In developed countries, the rate of washed coal burning for power generation can reach 60−100%.[36] However, in China, the proportion was less than 2%. With the propagation of national energy conservation policies, the proportion of coal washing in China is set to gradually increase over the next decade. As a result, an increasing number CFB boilers are expected be built to burn the vast amount of coal gangue produced by coal washing. In the near future, CFB boilers will need to be equipped with WFGD and SCR/SNCR setups for SO_2 and NO_X removal, which have the cobenefit of Hg removal.

SI Figures S3 and S4 illustrate the composition by province of the number of boilers and the mercury emissions of different-sized units in 2015, as summarized from Figures 4 and 5. Shandong had the most power plants but relatively low emissions and is thus ripe for expanding the policy of "Substitution of Smaller Units with Big Ones". Inner Mongolia had the most mercury emissions but fewer power plants than Shandong, Jiangsu and Zhejiang. Expanding on Figures 4 and 5, SI Figure S5 magnifies the geographical distribution of Mercury emissions from PC and CFB boilers for the Beijing-Tianjin-Hebei region, the Yangtze River Delta region and Guangdong in 2015.

3.2. Temporal Variation of Mercury emissions. Chemistry and transport models require the input of hourly mercury emission data. Accurate hourly and seasonal mercury emission data further our understanding of atmospheric Hg pollution. The monthly coal combustion for electricity generation was similar from 2013 to 2015 (SI Figure S6). In this article, we assumed that the mercury emission from a coal-fired power plant was proportional to the electricity generation. Figure 6 illustrates monthly variations by region (provincial variations are given in SI Table S6).

Overall, temporal variations in Mercury emissions were closely related to seasonal variations in industrial activities and wide fluctuations in the ambient temperature. Industrial and domestic electricity demands peak in summer and winter, which result in increased coal consumption and subsequent mercury emission. The highest coefficients occurred in December, corresponding to high year-end industrial activities.

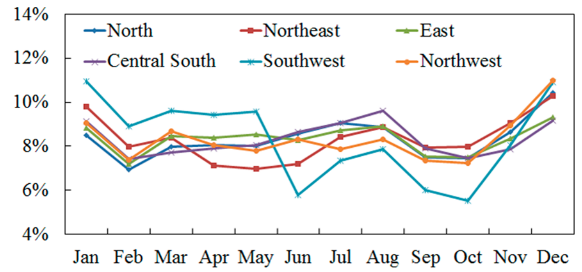

Figure 6. Monthly partition coefficients of regional Mercury emissions, 2015.

The lowest emissions in most regions (except the southwest region) occurred in February, depending on the timing of the Spring Festival. However, for southwest China, the monthly partition coefficients were obviously lower than those of other regions from June to August, and the lowest point occurred in October. As illustrated in SI Figure S7, hydropower accounted for over 35% of electricity generated in the southwest provinces in 2015. In Sichuan and Yunnan, in particular, this proportion was up to 85% and 86%, respectively. SI Figure S8 illustrates monthly variations in hydropower generation in the southwest provinces, showing a peak in summer. Therefore, the lower monthly partition coefficients of mercury emission in southwest China from June to August can be mainly attributed to the high proportion of hydropower generation during the summer.

According to statistics from the State Grid Corporation of China, there is little variation in the average load power for each day of the week, as shown in SI Figure S9. However, there are large fluctuations in the hourly partition coefficients of electricity generation in summer and winter. As illustrated in SI Figure S10, the hourly partition coefficients were higher during the time at which people are most active, from 8:00 to 21:00. The energy demand and corresponding mercury emission peaked early at 11:00 in the morning and again between 15:00 and 18:00 in the afternoon.

3.3. Mercury Emission Intensity. Due to the differences in coal consumption, Hg contents in the feed coals, coal types, and APCD types in coal-fired power plants, remarkable inconsistencies are observed in the mercury emission intensity (Hg-intensity) among the provinces−the lowest was 12.8 g/GWh in Beijing, and the highest was 33.7 g/GWh in Chongqing. In the U.S., the mercury emission limit for new or restructured low-rank coal-fired units is 0.04 lb/GWh, equivalent to 18.1 g/GWh.[59,60] The average Hg-intensity over the Chinese mainland is 18.3 g/GWh, which is close to the limit for low-rank coal-fired units in the U.S. In addition, 14 provinces had Hg-intensities less than 18.1 g/GWh. However, the mercury emission limit for active high-rank coal-fired units in the U.S. is 0.013 lb/GWh (equivalent to 5.9 g/GWh), which is much lower than the average Hg-intensity over the Chinese mainland.[59,60] See Table 2.

In 2015, the average Hg removal efficiency of APCDs was 75% in Chinese coal-fired power plants. If anthracite-fired power plants met the limit of 5.9 g/GWh and others met the limit of 18.1 g/GWh, the national average Hg removal efficiency of APCDs would improve to 88%. If all of the APCD removal efficiencies reached 92%, all Chinese coal-fired power plants could attain the 5.9 g/GWh standard. Both types of APCDs for ultralow emission—SCR+ESP-FF+WFGD and SCR+ESP+WFGD+WESP—can achieve a mercury removal efficiency of 92% in PC boilers. Consequently, Chinese coal-

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Table 2. Mercury Emissions from Coal-Fired Power Plants in China, 2015

	Hg emission (t/yr)				Hg-Intensity		Hg emission (t/yr)				Hg-intensity
province	total	Hg^0	Hg^{2+}	Hg_P	g/GWh	province	total	Hg^0	Hg^{2+}	Hg_P	g/GWh
Beijing	0.08	0.06	0.02	0.00	12.81	Hubei	2.04	1.56	0.47	0.01	22.30
Tianjin	0.67	0.47	0.19	0.01	12.86	Hunan	1.47	1.08	0.37	0.01	22.06
Hebei	3.55	2.46	1.02	0.05	16.42	Guangdong	3.55	2.56	0.94	0.05	12.86
Shanxi	3.66	2.69	0.94	0.03	16.60	Guangxi	0.87	0.66	0.20	0.00	17.71
Inner Mongolia	9.65	7.16	2.39	0.11	29.66	Hainan	0.19	0.14	0.05	0.00	13.04
Liaoning	2.50	1.87	0.61	0.02	19.42	Chongqing	1.41	1.08	0.32	0.01	33.69
Jilin	1.46	1.03	0.42	0.02	26.80	Sichuan	1.25	0.96	0.28	0.01	29.64
Heilongjiang	1.23	0.89	0.33	0.01	16.73	Guizhou	1.80	1.39	0.40	0.02	19.28
Shanghai	1.16	0.88	0.28	0.00	15.56	Yunnan	0.64	0.50	0.13	0.00	25.11
Jiangsu	7.25	5.50	1.71	0.04	18.81	Xizang	0.00	0.00	0.00	0.00	0.00
Zhejiang	3.26	2.47	0.77	0.01	15.41	Shaanxi	3.32	2.56	0.74	0.02	24.07
Anhui	3.75	2.73	1.00	0.03	20.14	Gansu	0.95	0.72	0.22	0.01	13.86
Fujian	1.89	1.46	0.42	0.01	18.37	Qinghai	0.23	0.18	0.05	0.00	19.83
Jiangxi	1.44	1.05	0.38	0.01	19.39	Ningxia	2.07	1.60	0.46	0.01	21.42
Shandong	7.55	5.69	1.79	0.06	17.62	Xinjiang	1.09	0.79	0.29	0.02	12.99
Henan	3.10	2.22	0.83	0.06	13.15	total	73.08	54.41	18.02	0.64	18.28

fired power plants could reach the emission limit for high-rank coal-fired units in the U.S. (5.9 g/GWh) in the near future with the implementation of the "Ultra-Low Emission and Energy Saving of Coal-fired Power Plant Plan".

3.4. Comparison with Other Studies. A series of articles have reported mercury emissions from coal-fired power plants, which were calculated to be a major contributor of anthropogenic Mercury emissions in the Chinese mainland (SI Figures S11−S12).[6−8,40,61,62] SI Figure S12 shows a comparison of provincial mercury emissions in 2010, determined by the methods in Zhang et al. (2015) and those in this study.[7] National emissions reached 100 tons in Zhang et al. (2015) but were estimated to be 109 tons in our method by considering the effects of boiler type and coal type. In addition to the difference of total emissions, the provincial distribution presented a significant difference. For example, mercury emissions in Inner Mongolia determined by this study's method were 3 tons higher compared with that by Zhang et al. (2015),[7] which was because 48% of the feed coal for PC boilers was lignite coal and the corresponding removal efficiencies were lower than those for other types of coal. Another reason was that CFB boilers burning coal gangue were calculated separately, which were ignored in Zhang et al. (2015).[7] Mercury emissions in Guizhou determined by this study's method were 1 ton lower, because 79% of the feed coal was anthracite and the corresponding removal efficiencies were higher than those for other types of coal.

SI Figure S13 shows variations in the special distribution determined by Streets et al. (2005),[14] by Zhang et al. (2010)[21] and in this study. From 2010 to 2015, with the implementation of national energy conservation policies, many small PC units were substituted by big units, especially in the Beijing-Tianjin-Hebei region and the Yangtze River Delta region. In contrast, a few new CFB boilers were established in Inner Mongolia and Shanxi for the effective utilization of low-grade coals. This is the first study to develop a monthly mercury emission inventory from coal-fired power plants at the province level. Wang et al. (2014)[57] simulated the surface concentration of East Asia using the anthropogenic inventory of Zhang et al. (2015),[7] while we simulated it with the highly resolved mercury emission inventory.[57] In the study of Wang et al. (2014)[57], the seasonal trend in the simulated Hg^0 concentration was opposite to the observations at Miyun station in north China in 2009. SI Figure S14 shows the comparison of simulated and observed Hg^0 at Mt. Changbai station in north China in 2015. The model accurately predicted seasonal variations, but the simulations were somewhat lower mainly due to the underestimation of natural source emissions.[63]

3.5. Uncertainty Analysis. Monte Carlo simulations were used to analyze the uncertainty in the total Mercury emissions. Because the coal consumption data were obtained from the China Energy Statistical Yearbook and the China Editorial Power Industry Statistics, a normal distribution with a coefficient of variation (CV, the standard deviation divided by the mean) of 5% was assumed for coal consumption. The Hg content was fitted with the log-normal distribution curve, as detailed in Zhang et al. (2015).[7] The distribution characteristics of the APCD removal efficiencies are shown in SI Table S4.[15,19] The overall uncertainty (95% confidence interval around the arithmetic mean) in the atmospheric Mercury emissions from Chinese coal-fired power plants in 2015 was estimated to range from −19% to 20%. Therefore, the variation in the Hg content in coal was the major contributor to the uncertainty, closely followed by the APCD removal efficiency. Based on current practices, further reduction of the uncertainties arising from the Hg content in coal is not practical. The APCD mercury removal efficiency could be further reduced by attritional field testing for ultralow emission devices.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b06209.

Tables S1−S6 and Figures S1−S14 (PDF)

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Notes

The authors declare no competing financial interest.

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