Atmospheric mercury emissions from coal-fired power plants in Northern China

ATMOSPHERIC MERCURY EMISSIONS FROM COAL-FIRED POWER PLANTS IN NORTHERN CHINA

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ABSTRACT

Hg emissions from coal-fired power plants (CFPPs) in Northern China are the major source of anthropogenic atmospheric mercury (Hg) emissions. The impact of denitrification devices on Hg emission, together with difference in Hg emission from different boilers and specific data of air pollution control devices (APCDs) are critical factors for the estimate of Hg emissions. Combined with unprecedented meta-analysis, this paper applies emission factor method to examine Hg emissions in 1995, 2003, 2011 and projection to 2015. Hg emissions from CFPPs were estimated to rise from 13.28 ton to 23.61--36.67 ton with an increase rate of 77.8% to 176.3%, while coal consumption increased by 325.5%. The emissions of Hg^0, Hg^{2+} and Hg_P are estimated to reach 11.80--18.34 ton, 9.45--14.66 ton and 2.36--3.67 ton, respectively. Low increase of Hg emissions benefits from the wide application of APCDs, especially the sound operation of denitrification devices. However, there are uncertainties in the estimation of Hg emission due to variation of parameters in the method, and Hg content was proved to be the predominant parameter that results in the uncertainties. The paper addresses the effect of denitrification devices and their operation rate on Hg removal, and reduces the uncertainties of Hg emissions through comprehensive emission factors.

KEYWORDS: coal-fired power plant; mercury emissions; emission factor method; uncertainty

1. INTRODUCTION

Atmospheric Mercury (Hg) emission from coal-fired power plants (CFPPs) has been the central environmental issue for Northern China since the release of 12th five-year plan of China in 2011. Hg is a naturally occurring metal and has aroused concern owing to its toxicity, long-range transportation, persistence and bio-concentrate in environment. Coal combustion is generally regarded as the significant source of Hg emission for the global inventory. Further, the major utilization of coal is to generate power [1, 2]. This trace metal is released and distributed in fly ash, gypsum and bottom ash in CFPPs (Fig. 1), during coal combustion in the form of Hg^0, Hg^{2+} and Hg_p [3, 4]. Also it is vaporized into flue gas [5, 6].

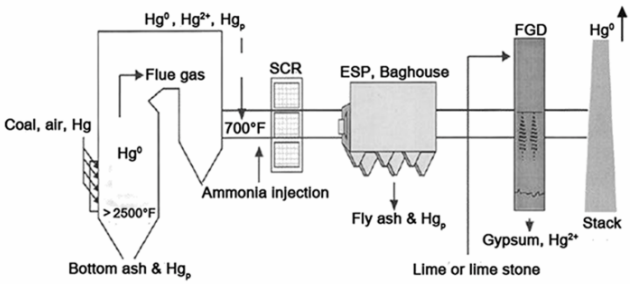

FIGURE 1 - Hg distribution in coal combustion. Hg distributes in bottom ash, fly ash and gypsum in the process of coal firing. The speciation of Hg in combustion includes Hg^0, Hg^{2+}, Hg_p.

Hg removal from flue gas is usually gained through measures aimed to remove conventional contaminants in exhaust. United States Environmental Protection Agency (EPA) reported the air pollution control devices (APCDs), installed for removing NO_x, SO_2 and particulate matter, can achieve the co-benefits of Hg capture, especially Hg^{2+} [7]. Field-test data shows that Hg control can be achieved in coal-fired unit equipped with flue gas desulphurization (FGD) for SO_2 control, bag-house filter or electrostatic precipitator (ESP) for particulate matter (PM) control, and denitrification devices, e.g. selective catalytic reduction (SCR) for NO_x emission control [8]. Nevertheless, the impact of denitrification devices on Hg removal has been seldom involved in the estimation of Hg emissions regarding China owing to the difficulty in acquiring data of denitrification rate, since the capacity of CFPPs installed with SCR before 2010 is very small and some of them are suspected not to put into full operation due to additional operation cost, e.g. catalyst and injection of NH_3 [9].

It is estimated that total anthropogenic Hg emissions were 2320 ton for 2008, and coal and oil combustion are

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identified as the most significant anthropogenic sources - 35% of total anthropogenic emissions [10]. Moreover, China is the largest Hg emitter in the world and CFPPs have become a dominant source of anthropogenic atmospheric Hg emissions and exposure [2, 11]. This is especially the case in Northern China, where CFPPs assume a role in both generating electricity all over the year and supplying heat power for residents in winter, and providing steam for factories as well (Fig. 2).

FIGURE 2 - The geographical range of research area. Research area is 1, 556, 000 km^2 and constitutes Beijing, Hebei, Inner Mongolia, Shanxi and Tianjin.

Since CFPPs are the major anthropogenic source of Hg emission in some countries, e.g. China, Poland and Southern Africa; much effort has been made to study and control regional Hg emission from CFPPs by researchers and authorities [12-15]. The usual method used to estimate Hg emissions from CFPPs is on the basis of emission factors, with regard to Hg content in coal, types of boilers and Hg removed by coal washing and emission control devices. Nonetheless, most of previous studies, which carried out to estimate Hg emissions from CFPPs, lack comprehensive consideration in emission factors. For example, Dabrowski et al. [13] and Glodek and Pacyna [12] did not involve the emission factor of boilers at each CFPP for their method. Wu et al. [15] and Tian et al. [16] ignored the effect of denitrification devices on Hg emissions. Wang et al. [17] did not estimate Hg emissions from CFPPs on the grounds of specific data of APCDs installation for each CFPP. Zhang et al. [18] did not consider difference between the boilers of CFPPs and industrial boilers or domestic boilers in evaluating Hg emissions from coal combustion. Due to the distinguished and incomplete consideration in the above studies, their result could hardly provide reliable clue for the trend of Hg emissions and valuable perspective for policy making.

Apart from the limit of current studies of Hg emissions, the estimates of Hg emission are prone to generate large uncertainties, both in the total amount of Hg emissions and speciation. Hg speciation is significant in controlling atmospheric and deposition processes, and thus the fate of Hg in the environment [19].

In this paper, we present a comprehensive estimation of Hg emissions from CFPPs in 1995, 2003 and 2011 in Northern China, and then project to the future and estimate Hg emissions, including Hg speciation, in 2015. 1995, 2003 and 2011 were chosen because Emission Standard of Air Pollutants for Thermal Power Plants witnessed three revisions and implemented in the next year after the three years. Particularly, to get best Hg estimate, unprecedented meta-analysis was conducted concerning emission factor of different boilers, the installation rate of APCDs, and Hg removal efficiency and the operation rate of denitrification devices. In the pages that follow, it is argued that uncertainties cannot be neglected in the estimation of Hg emissions due to the variation of estimation parameters.

2. MATERIALS AND METHODS

A bottom-up method of emission factors was adopted in this study to estimate atmospheric Hg emissions from coal-fired boilers of power plants. Hg emissions from CFPPs were calculated by combining the detailed data of coal consumption, the averaged Hg content in coals by province and the specific emission factors that are classified by different boiler patterns, and PM, SO_2 and NO_X control device configuration. The basic formulas can be expressed as follows.

$$\mathbf{E} = \sum_{i}\sum_{j}\left[M_i A_{i,j}(\mathbf{1} - Q.w)R_j\left(\mathbf{1} - \sum_{j}\sum_{k} P_{j,k}\,\boldsymbol{\eta}_{j,k}F_j\right)\right]$$

Where E is the Hg emissions from CFPPs, ton/year; M is the Hg content of coal as burned, mg/kg; A is the amount of coal consumption, 10^6 ton/year; Q is the percentage of washed coal in the power plants; w is the Hg removal efficiency of coal washing; R is the release factor of Hg from boiler; P is the application rate of a certain combination of APCDs; η is the Hg removal efficiency of one combination of APCDs; F is the operation rate of denitrification devices; i is the province; j is the combustor type; and k is the type of APCD combinations.

2.1 Coal transport and consumption

Coal consumption data of power plants were obtained from China Energy Statistics Yearbook [20-22]. Coal consumption in 1995, 2003 and 2011, including washed coal consumption, is shown in Fig. 3.

The research area is 1, 556, 000 km^2 and consists of Beijing, Tianjin, Hebei, Shanxi and Inner Mongolia. The provinces with large coal consumption do not have large coal production. Hence, there is interprovincial coal

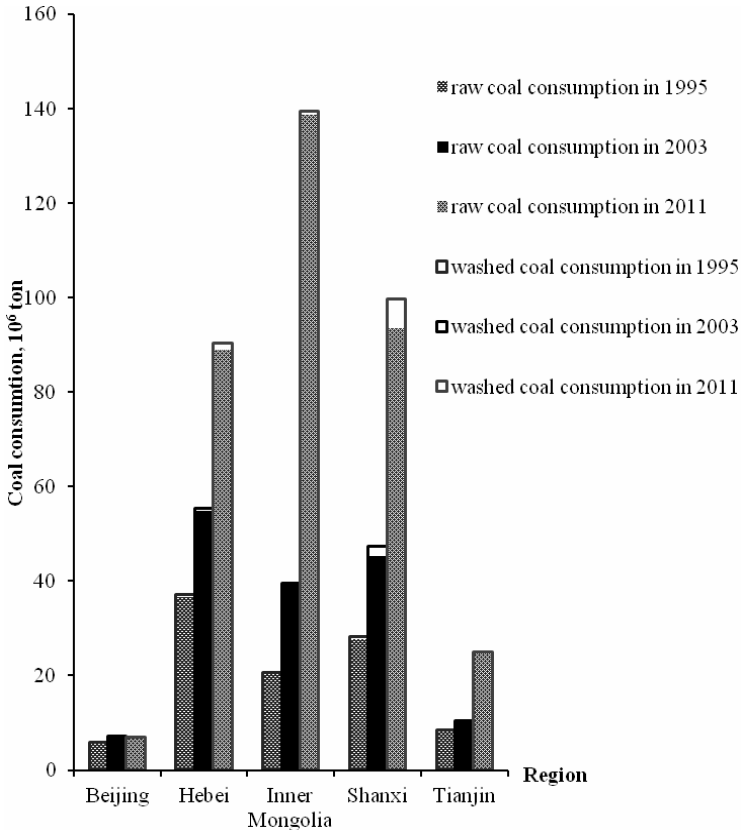


FIGURE 3 - Coal consumption at CFPPs during 1995 and 2011. Coal consumption at CFPPs in Inner Mongolia experienced the largest increase, while that in Beijing had minimum variation during 1995 and 2011. This implicates the difference in Hg emissions from different regions. CFPPs refer to coal-fired power plants. Total coal consumption constitutes raw coal and washed coal consumption.

transport. Coal in Shanxi and Inner Mongolia can satisfy their own need and the rest exports to other provinces, such as Beijing, Tianjin and Hebei. Coal transport can be described by developing a coal transport matrix [11].

$$m_c = Am_p$$

$$m_c = [m_{c1}, m_{c2}, \dots, m_{cn}]^T$$

$$A = \{a_{ij}\}n \times n$$

$$m_p = [m_{p1}, m_{p2}, \dots, m_{pn}]^T$$

Where vector m_c is the Hg content of coal as consumed in all the provinces; m_p is the Hg content of coal as produced in all the provinces; A is the coal transport matrix, and a_{ij} is the amount of coal transported from province j to province i, the sequence of each province in row and column is Beijing, Hebei, Inner Mongolia, Shanxi and Tianjin; n is the number of provinces.

$$A_{2011} = \begin{bmatrix} 1.10 & 0.98 & 0.00 & 4.88 & 0.00 \\ 0.00 & 20.01 & 35.13 & 35.13 & 0.00 \\ 0.00 & 0.00 & 136.50 & 3.03 & 0.00 \\ 0.00 & 0.00 & 0.79 & 98.89 & 0.00 \\ 0.00 & 3.13 & 0.00 & 21.86 & 0.00 \end{bmatrix}$$

$$A_{2003} = \begin{bmatrix} 2.17 & 0.83 & 0.00 & 4.13 & 0.00 \\ 0.00 & 12.30 & 21.60 & 21.60 & 0.00 \\ 0.00 & 0.00 & 38.63 & 0.86 & 0.00 \\ 0.00 & 0.00 & 0.38 & 46.99 & 0.00 \\ 0.00 & 1.32 & 0.00 & 9.21 & 0.00 \end{bmatrix}$$

$$A_{1995} = \begin{bmatrix} 0.30 & 0.93 & 0.00 & 4.66 & 0.00 \\ 0.00 & 8.26 & 14.50 & 14.50 & 0.00 \\ 0.00 & 0.00 & 20.24 & 0.45 & 0.00 \\ 0.00 & 0.00 & 0.22 & 27.92 & 0.00 \\ 0.00 & 1.07 & 0.00 & 7.47 & 0.00 \end{bmatrix}$$

2.2 Average content of Hg in raw coals as produced and consumed

The content of Hg in coal can provide useful information for pollution control during coal combustion and utilization from an environmental point of view. Even when being presented in only parts per million levels in coal, combustion of coal can result in tons of hazardous air pollutants being discharged into atmosphere. The content of Hg in coal mined from different places varies substantially due to the coal-forming geological environments. Previous studies demonstrated that the content and distribution of Hg differ from provinces, sources and even the coals of the same seam [23]. On top of the modes of occurrence, mineral content, and the distribution of Hg in coals, there are also some other sources causing fluctuation of hazardous trace elements contents, such as random sampling errors, measurement errors, or samples are not representative.

In this study, the tested content of Hg in raw coals was compiled and summarized from available published literatures, regarding research area. In the early studies, Hg content data were quite limited. Wang et al. [17], Wang [24] and Zhang et al. [18] used 0.22mg/kg as a national mean value, which was derived from coal analysis data of 14 provinces. The values varied from around 0.02 mg/kg to 1.92mg/kg. Other research yielded estimated values of 0.15 mg/kg [25] and 0.16 mg/kg [26]. All of these results came from very limited raw coal samples of coal mines. To develop a more convincing inventory, the U.S. Geological Survey (USGS) did further studies after analyzing 276 samples from all provinces in China and obtained an average Hg content of 0.15±0.14mg/kg. On the basis of data from USGS and other research, Streets et al. [27] presented a complete Hg content database by province in China. They also developed a coal transport matrix for China, combining coal production with coal consumption, and obtained a value of 0.19 mg/kg for the Hg content of coal burned in the power sector. Zheng et al. [28] analyzed 62 samples, summarized 1699 samples from previous studies, and reported the national average to be 0.19 mg/kg. Ren et al. [29] conducted a more detailed data investigation and outlined previous results of 619 samples in their book. The lognormal distribution was found to fit the input dataset of Shanxi province. The values with a probability of 50% for this province were more applicable than the mean values. The study presented here integrated the independent data from Ren et al. [29] with other research and updated database of Hg content in coal by province (Table 1).

TABLE 1 - Hg content in raw coal in research area, mg/kg

Region	This study	Ren et al. (2006) [29]	Wang et al. (2010) [30]	Streets et al. (2005) [27]	Zheng et al. (2007) [28]	Zhang et al. (2012) [2]	Tian et al. (2010) [23]
Beijing	0.33	0.10	0.55	0.44	0.34		0.34
Hebei	0.16	0.16(33)	0.15(14)	0.14		0.17(15)	0.13
Inner Mongolia	0.18	0.17(14)	0.17(15)	0.22	0.16	0.18(46)	0.20
Shanxi	0.17	0.17(79)	0.16(69)	0.16	0.08		0.17
Tianjin	-						

Data in the bracket are numbers of samples. Tianjin does not produce raw coal and so no numbers are given in the table.

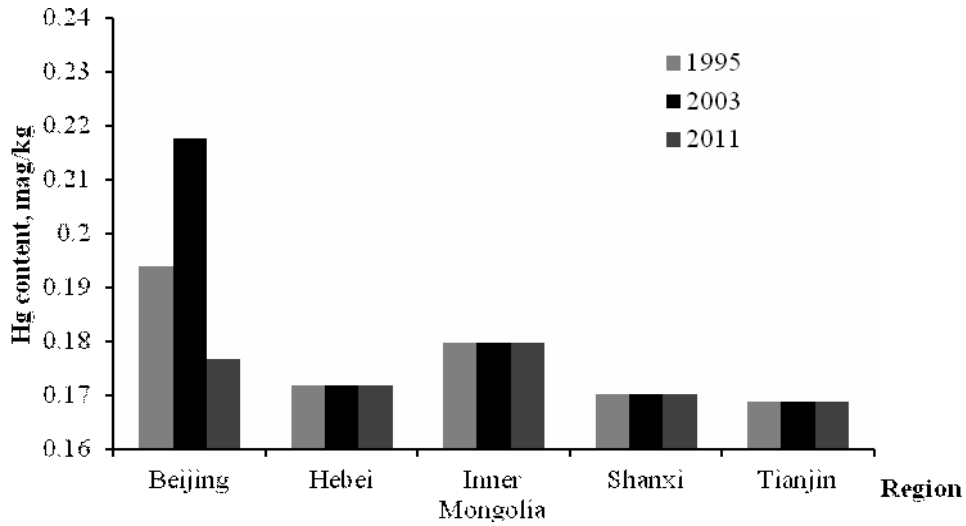


FIGURE 4 - Hg content in coal as consumed at CFPPs. Hg content in coal as consumed in Beijing showed fluctuation in 1995, 2003 and 2011. The variation derived from the difference in amount of coal import and large gap of Hg content between native coals and alien coals. The particularly high Hg content in 2003 resulted from combined high Hg content of native raw coals and more native coals consumed. The labels designate Hg content of coal used at CFPPs in 1995, 2003 and 2011, respectively.

For the major coal-supplying provinces such as Shanxi and Inner Mongolia, there is little difference between Hg content as produced and as burned in power plants because within-province supply can meet the demand. However, for the major coal-consuming regions such as Tianjin, the content of Hg that burned depends on coal from other provinces. The Hg content of coal used in power plants by province, as shown in Fig. 4, was calculated via the coal transport matrix and Hg content in raw coal of coal-supplying provinces. These values were all of the weighted means on the basis of the coal consumption by province.

2.3 Removal efficiency of Hg through APCDs

Aside from the Hg content in coal, Hg removal efficiency of APCDs is another critical parameter for the calculation of Hg emissions because they are effective in decreasing the final Hg release into atmosphere [30, 31], and have co-benefit effects of Hg speciation by SCR. For instance, Wang et al. [32] investigated Hg emissions from six CFPPs, and found the release rate and speciation of Hg varied substantially for different boilers and equipped PM and FGD systems. The observation of Laudal et al. [33] at seven CFPPs indicated that SCR can assist in converting Hg^0 to Hg^{2+}, thereby reducing the final emissions of elemental Hg into the atmosphere. In our study, CFPPs in research area were classified into various subgroups based on boiler patterns, particulate control devices, FGD systems and NOx reduction devices, which were seldom considered in the existing studies of Hg emissions from CFPPs in China. The SCR catalyst can alter the speciation of Hg and promote some of elemental Hg to oxidize into Hg^{2+}, which is much easier to be absorbed by the downstream wet FGD scrubber.

Table 2 demonstrates Hg removal efficiencies for overall 8 APCDs and their combinations. The results of 107 samples were assembled from existing studies involved in China, America, Japan and Korea. PC +ESP and PC+ESP+WFGD are two important APCDs combinations in the research area at different phases of our research. Furthermore, PC+ESP+WFGD+SCR have been increasing at a staggering rate throughout the research area since 2011, in order to comply with the strict Emission Standard of Air Pollutants For Thermal Power Plants (GB13223-2011) [47]. There are enough data for the former two combinations, 35 results for PC+ESP and 24 results for PC+ ESP+WFGD.

SPSS16.0 and Oracle Cristal Ball 11.1.2.2 were used to find a statistical distribution to fit these data. As a con-

TABLE 2 - Hg removal efficiencies of APCDs

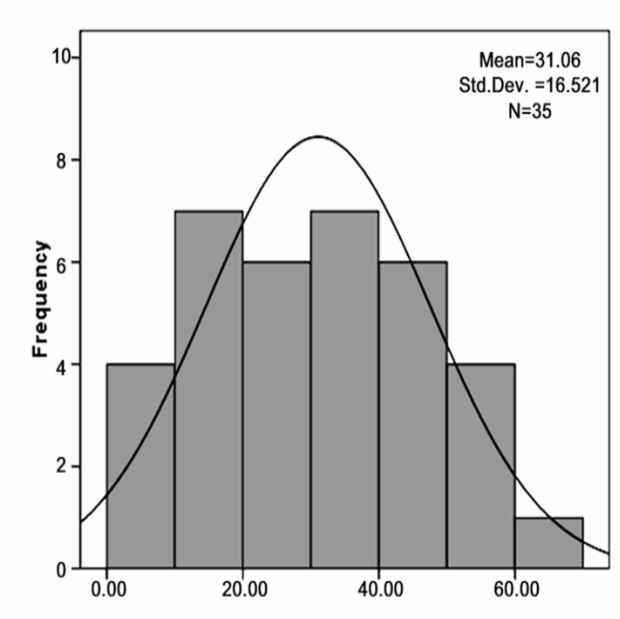
Control device	Numbers of samples	Removal efficiencies, %			
		P10	P50	P90	Mean
ESP	35[a]	7.38	30.20	53.28	31.06
FF	15[b]	43.20	84.00	99.00	75.57
PCRB	8[c]	4.30	15.00	46.00	18.25
CYC	2[d]	0.00	0.05	0.20	0.05
WFGD	11[e]	17.06	60.00	79.00	55.98
PC+ESP+WFGD	24[f]	22.20	69.00	78.00	59.92
CFB-FGD+ESP	6[g]	38.00	68.50	100.00	71.67
SCR+ESP+WFGD	6[h]	36.00	80.05	95.00	75.97

a. [4, 16, 32, 34-40]; b. [4, 16, 35, 37, 39, 40]; c. [4, 16, 35, 38]; d. [38, 40]; e. [6, 16, 39, 41-44];
f. [4, 32, 35-40]; g. [2, 4, 40, 45]; h. [2, 32, 36, 37, 44, 46]; i. PC refers to pulverized-coal. FF refers to fabric filter; PCRB refers to PM scrubber; CYC refers to cyclone; CFB-FGD refers to circulating fluidized bed FGD. WFGD refers to wet FGD.

TABLE 3 - APCDs proportion of CFPPs in 1995, 2003 and 2011

Region	Control factor	1995	2003	2011
Beijing	PM	87.64%	100.00%	100.00%
	SO_2	0.00%	23.89%	100.00%
	NO_X	0.00%	0.00%	81.83%
Hebei	PM	91.86%	100.00%	100.00%
	SO_2	0.00%	1.66%	100.00%
	NO_X	0.00%	0.00%	8.37%
Inner Mongolia	PM	89.61%	100.00%	100.00%
	SO_2	0.00%	0.00%	100.00%
	NO_X	0.00%	0.00%	3.08%
Shanxi	PM	94.72%	100.00%	100.00%
	SO_2	0.66%	1.66%	100.00%
	NO_X	0.00%	0.00%	3.08%
Tianjin	PM	92.83%	100.00%	100.00%
	SO_2	0.00%	0.00%	100.00%
	NO_X	0.00%	0.00%	13.16%

a.



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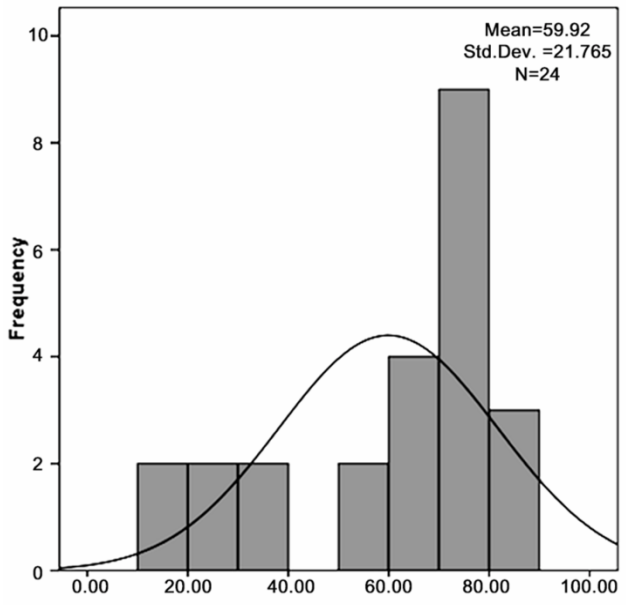

FIGURE 5 - Distribution curves of Hg removal efficiencies of APCDs. Hg removal efficiencies of APCDs (a. Hg removal efficiency of ESP and b. Hg removal efficiency of ESP+WFGD) are in line with Normal distribution and Weibull distribution, respectively. The mean value of ESP+WFGD deviates more from P50 because of the long tail of distribution curve, compared with that of ESP. N refers to numbers of samples; Std.Dev refers to standard deviance; Mean refers to arithmetic value of the samples.

sequence, the Hg removal efficiencies of PC+ESP and PC+ ESP+WFGD were in line with the Normal distribution and Weibull distribution, respectively (Fig. 5). The reported mean values of the two combinations are 29% and 62%, respectively [2]. The P50 value for the Hg removal efficiency of PC+ESP and PC+ESP+WFGD turned out to be 30.20% and 69%, higher than the mean values.

Through collecting data from the national inventory of desulfurization and denitrification facilities [48] and China Energy Yearbook [21, 22, 49], the capacity of units equipped with APCDs in 1995, 2003 and 2011 was calculated and installation rate of different APCDs at each CFPP in research area was analyzed (Table 3).

2.4 Emission rate of Hg for combustion devices

Combustion devices that are used at CFPPs are divided into three types: PC boilers, CFB furnaces, and stoker-fired boilers.

PC boilers have accounted for a large proportion at CFPPs since 1995. The remaining has been shared by the other two patterns, which have been commonly adopted in small units. The national average proportion of installed capacity for PC boilers, CFB furnaces, and stoker-fired boilers are roughly 83.3%, 12.9%, and 3.8%, respectively [50]. Some researchers, abroad and domestic [31, 32, 51-55] investigated Hg release rate for various types of boilers at CFPPs (Table 4) and they found over 99% of Hg in coals will evaporate into flue gas. Here, the arithmetic mean value of Hg emission ratio was adopted for different types of utility boilers reported in available literature, and we assume that the averaged emission ratios of Hg were about 99.5% for pulverized-coal boilers, about 83.2% for stoker-fired boilers, and approximately 98.9% for fluidized-bed combustion boilers, respectively.

TABLE 4 - Hg release rates of different boiler types

Boiler type		Release Rate, %	Reference
PC boiler		99.50	[54]
PC boiler		99.53	[56]
PC boiler		99.10	[55]
PC boiler		99.13	[43]
PC boiler		99.77	[31]
PC boiler		99.73	[53]
PC boiler		99.78	[32]
In this study	Stoker fired boiler	83.15	[57]
	CFB furnace	98.92	[51]
	PC boiler	99.47	Mean value

2.5 Removal efficiency of Hg through coal washing

Currently, coal washing before combustion is an effective way to reduce ash and SO_2 emissions. It can reduce sulfur pyrites content by 40% [58]. This way, not only diminishes ash and SO_2 emissions, but also decreases Hg concentration and increases the heating value of cleaned coal. The removal effect of Hg is approximately similar to the sulfur in pyrite and it is indicated that sulfur in pyrite can be removed effectively during physical coal cleaning and the removal efficiency can reach up to 50%

[59, 60]. Consequently, a large proportion of Hg associated with pyrite can be washed away simultaneously. The reported Hg removal efficiency of coal cleaning varied from about 30% to 75% and all the physical coal cleaning procedures tested on coals can remove larger than 50% Hg [8, 59-62]. In this study, we assumed a mean removal efficiency of 50%.

Although APCDs occupied a considerable proportion at CFPPs in research area and the share has risen at a staggering rate, washed coal only accounted for 2.2% and 2.7% of the electricity coal consumption in China in 2003 and 2011, respectively [15, 49]. However, as is shown in Fig. 4, it was quite varied in different phases in research area.

2.6 Operation rate of denitrification devices

Even though denitrification devices are highly efficient in removing Hg, the effect of Hg removal has been offset by the low operation rate in China. Before denitrification electricity price is implemented in 2013, the operation rate of denitrification devices was investigated by China Electricity Council (CEC) to be 17% [63]. In this study, 17% was adopted as the value of F to estimate Hg emissions in 2011 and ignored in 1995 and 2003, as the installation rate of SCR or SNCR was negligible before 2011 for research area.

2.7 Hg speciation

Hg is mainly released to the air as gaseous Hg^0, and also mixing gaseous Hg^{2+} and particulate Hg_p at CFPPs. Hg speciation was estimated from the global technical report for evaluating Hg emissions [64]. Specifically, 'split factors' of 0.5, 0.4 and 0.1 were applied to Hg^0, Hg^{2+}, Hg_p split, respectively.

3. RESULTS AND DISCUSSION

3.1 Hg Emissions in 1995, 2003 and 2011

A provincial inventory of Hg emission for CFPPs is indicated in Table 5. Hg emissions were 13.28 ton, 18.81 ton and 22.71 ton in 1995, 2003 and 2011, respectively. As can be inferred from recent research regarding research area, Hg emissions ranged from 19.28 ton to 26.50 ton in 1995 [17, 18, 38].

The difference of Hg emissions from CFPPs in research area between other studies and this study is largely owing to a much coarser data sources or a much simpler method in other studies. Zhang et al. [18] applied Hg emission ratio, which did not differ the boilers of CFPPs from industrial boilers or domestic boilers in evaluating Hg emissions from coal combustion. Wang et al. [17] and Wu et al. [38] applied the same method as our study, but they adopted a statistic average of CFPPs installation rate rather than acquiring data from each CFPP in prediction of Hg emissions, and ignored the influence of denitrification devices on Hg removal. In contrast, we conducted meta-analysis of installation rate for APCDs from each CFPP and involved the impact of denitrification devices on Hg emission. Accordingly, it can be convincingly argued that the estimation of Hg emissions presented in this study is more accurate.

The top emitter in 1995 and 2003 was Hebei, whereas Hg emissions of Inner Mongolia in 2011 almost doubled in comparison with that in 2003, and became the top emitter. The top emitters in the three years contributed 36.63%, 34.07% and 40.19% of the total Hg emissions from the power sector in research area. From 1995 to 2011, coal consumption increased by 259.6%, which means the total national Hg emissions rise by 259.6%. If no specific control strategies were implemented, in fact, the total emissions would rise from 22.60 ton to 58.67 ton because further SO_2 and NO_x control measures counteracted most of the influence brought about by the rapid growth of coal consumption. There is a reduction of 35.96 ton with respect to Hg emissions during the 16 years, which attributes to progress in desulfuration and denitrification.

The effects of control strategies for SO_2 and NO_X in each province were quite different. Inner Mongolia saw a dramatic increase in Hg emissions at CFPPs from 1995 to 2011 (+215.9%), whereas Beijing witnessed obvious decline in Hg emissions during this period (-56.2%). The discrepancy partly derived from quick increase of coal consumption in electricity generation from Inner Mongolia. Furthermore, APCDs, particularly SCR, FGD and their combinations with ESP and FF, were more prevailed in Beijing than in other provinces. This may be the main reason that could explain the reduction of Hg emissions in Beijing.

3.2 Hg Emissions Projection to 2015

According to the circular of the State Council in China (SC), by 2015, coal consumption in CFPPs will be constantly rising at an annual rate of 4.3%, while coal washing rate will be over 65% [65]. Furthermore, all new-

TABLE 5 - Provincial Hg emissions at CFPPs in 1995, 2003 and 2011

Region	1995		2003		2011	
	Emissions, ton	Share, %	Emissions, ton	Share, %	Emissions, ton	Share, %
Beijing	0.80	6.05	0.97	5.14	0.35	1.55
Hebei	4.86	36.63	6.41	34.07	5.63	24.79
Inner Mongolia	2.89	21.77	4.83	25.67	9.13	40.19
Shanxi	3.62	27.27	5.39	28.67	6.02	26.51
Tianjin	1.10	8.28	1.21	6.45	1.58	6.95
Northern China	13.28		18.81		22.71	

TABLE 6 - Total Hg emissions and speciation for the research area

Scenarios	F	Total Hg emissions, ton	Hg speciation, ton		
			Hg^0	Hg^{2+}	Hg_p
(1)	17%	36.67	18.34	14.66	3.67
(2)	50%	31.48	15.74	12.59	3.15
(3)	100%	23.61	11.80	9.45	2.36

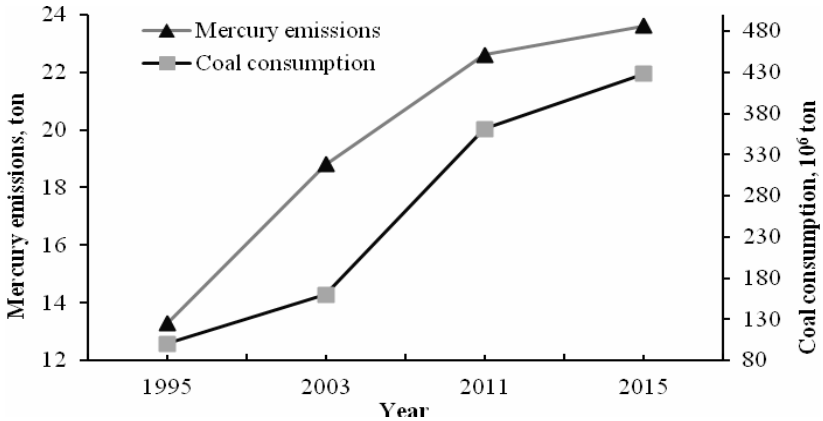

FIGURE 6 - Hg emissions at CFPPs during 1995 and 2015. The estimate for 2015 was based on the hypothesis that the operation rate of denitrification devices reaches 100%. Hg emissions at CFPPs experienced quicker increase than coal consumption from 1995 to 2003. The trend took a turn after 2003. The popularity and sound operation of APCDs are mainly responsible for the transformation. The labels designate mercury emissions and coal consumption, respectively.

built coal-fired units will be equipped with control facilities for SO_2 and NO_X [66].

We assume the above goals would be realized in the research area, and consider different scenarios, with respect to the operation rate of denitrification devices, i.e. 17%, 50% and 100% (Table 6). Therefore, Hg emissions from CFPPs in 2015 can be estimated and a figure of 23.61 ton was acquired for total Hg emissions when denitrification devices operate well with the units, and 11.8 ton, 9.45 ton and 2.36 ton for Hg^0, Hg^{2+} and Hg_P emissions, respectively. Combined with coal consumption, a value of 0.0552 to 0.0857 g Hg ton^{-1} coal for the research area was calculated under different scenarios. This value compares well with that of other regions, including the USA and Canada (0.0094 to 0.18 g Hg ton^{-1} coal) [67], South Africa (0.02 to 0.16 g Hg ton^{-1} coal) [13] and global estimates (0.04 to 0.3 g Hg ton^{-1} coal) [10].

As shown in Fig. 6, emissions experienced a steady increase rate of 3.96% from 2011 to 2015, while coal consumption went up rapidly by 18.34% during the same period. An overall increase rate of 77.8% for Hg emissions was calculated from 1995 to 2015. This result mainly bases on the assumption that SCR will be applied comprehensively and operate consistently and stably with the units at CFPPs in the 12th five-year

3.3 Uncertainties

Uncertainties widely exist in the estimation of Hg emissions. The common considerations underpinning the uncertainties involve limited measurements of Hg emission and the accuracy of emission factors [68, 69].

In order to examine uncertainties of Hg emissions, the Monte Carlo method was applied to conduct the stochastic simulations of Hg emissions in 2011. To get reliable outputs, we set the sampling number as 6000. All Hg emissions from the five regions are now shown by distribution curves instead of single values.

The curves demonstrate a wide range of uncertainties. As can be seen from Fig. 7, total Hg emissions from CFPPs in 2011 ranged from 3.4 ton to 36.6 ton, an order of magnitude difference. To determine which parameter is the largest source of the uncertainty, various scenarios were designed.

(1) A complete stochastic simulation with distribution functions regarding various parameters, including Hg content, Hg removal efficiency of APCDs and coal washing.

(2) Only set Hg content in coal with distribution function.

(3) Only set Hg removal efficiency of APCDs with distribution functions.

(4) Only set Hg removal efficiency of coal washing with distribution functions.

As can be seen from Table 7, Hg content in coal contributes the largest uncertainty of Hg emissions. The second largest source of uncertainties is Hg removal effi-

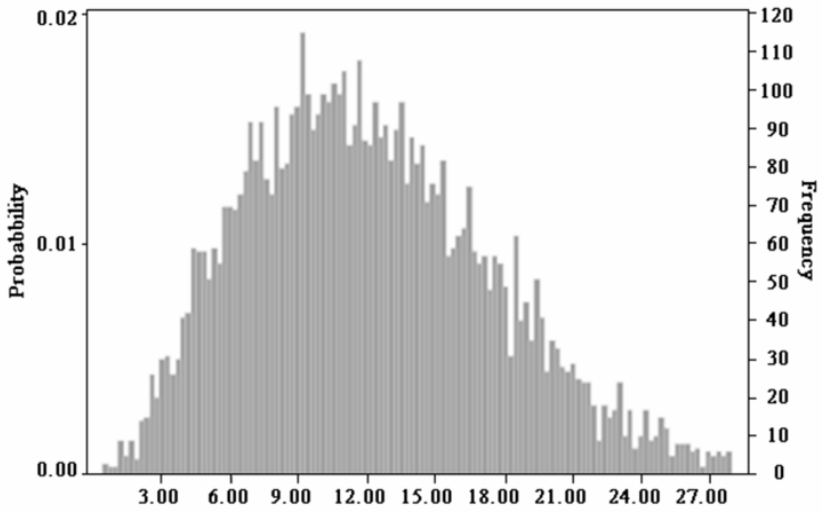

FIGURE 7 - The output distribution curves for Hg emissions in 2011 at CFPPs. Stochastic simulation of Hg emissions is conducted via the Monte Carlo method by Oracle Cristal Ball, with the sampling number 6000. Hg content variation is the predominant factor that contributes to the uncertainties of Hg emissions. The unit is ton.

ciency of APCDs, while Hg removal efficiency of coal washing has minimal impact on uncertainties of Hg emissions. Wu et al. [15] estimated an uncertainty range of -68.0% to 199.8% for Hg emissions, on the grounds of a complete stochastic simulation with all parameters built with distribution functions. Overall, this judgment is in agreement with scenario (1) of our study.

TABLE 7 - The uncertainties of Hg emissions with various scenarios setting

Scenarios	Hg missions P50, ton	Uncertainty range, %
(1)	14.7	-76.9 to +149.1
(2)	18.6	-74.6 to +124.5
(3)	18.2	-44.5 to +24.2
(4)	22.6	-0.6 to +0.7

4. CONCLUSIONS

The present study was designed to include comprehensive estimation of Hg emissions from CFPPs. Considering emission factor in coal cleaning, combustion and atmospheric pollutant removal, the best estimate for Hg emissions from CFPPs in 1995, 2003, 2011 in research area arrives at 13.28 ton, 18.81 ton, 22.71 ton, respectively, while that of 2015 ranges from 23.61 ton to 36.67 ton, subject to the operation rate of denitrification devices. Hg^0, Hg^{2+} and Hg_p are predicted to be 11.80-18.34 ton, 9.45 - 14.66 ton and 2.36-3.67 ton in 2015. However, Hg emissions in 1995 in the current study differ from the existing estimate of 19.28 ton to 26.50 ton. A possible explanation for the difference may be the more limited data sources or oversimplified estimation method in other studies.

During 1995 and 2015, Hg emissions from CFPPs were predicted to rise by 77.8% to 176.3% while coal consumption increased by 325.5%. We concluded that the slower increase rate of Hg emissions resulted from the wide use of FGD, SCR and their combination with PM control devices. To reach the slower increase of Hg emissions in the future, the popularity and operation rate of denitrification devices are the key. As desulphurization electricity price has been successfully carried out since the 11th five-year plan, likewise, denitrification electricity price, which is ruled by Chinese government in 2013, will impel the installation of denitrification devices [70, 71]. Nonetheless, the current policy should be completed by differentiating electricity and heat generators from electricity generators in electricity price.

Even though various possibilities were considered in the process of calculation, uncertainties cannot be ignored in the estimation of Hg emissions. The Monte Carlo method was applied to perform the stochastic simulation of Hg emissions in 2011. Consequently, uncertainties of Hg emissions ranged from -76.9% to 149.1% and Hg content was proved to be the predominant parameter contributed to the uncertainties. Although it is too early to draw statistically significant conclusions, the prediction seems to be more accurate with more field test data of coal samples and APCDs removal efficiency from different CFPPs in the future. Our conclusions should provide insight into Hg abatement for policy making, and be used in transport and fate modelling of Hg emissions.

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