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# Mercury removals by existing pollutants control devices of four coal-fired power plants in China

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#### Abstract

The mercury removals by existing pollution control devices and the mass balances of mercury in four coal-fired power plants of China were carried out based on a measurement method with the aluminum matrix sorbent. All the plants are equipped with a cold-side electrostatic precipitator (ESP) and a wet flue gas desulfurization (FGD) in series. During the course of coal stream, the samples, such as coal, bottom ash, fly ash, gypsum and flue gas, were collected. The Hg concentrations in coals were measured by CVAAS after appropriate preparation and acid digestion. Other solid samples were measured by the RA-915<sup>+</sup> Zeeman Mercury Spectrometer. The vapor phase Hg was collected by a sorbent trap from flue gas and then measured using CVAAS followed by acid leaching. The mercury mass balances were estimated in this study were 91.6%, 77.1%, 118% and 85.8% for the four power plants, respectively. The total Hg concentrations in the stack gas were ranged from  $1.56-5.95 \ \mu g/m^3$ . The relative distribution of Hg in bottom ash, ESP, WFGD and stack discharged were ranged between 0.110%-2.50%, 2.17%-23.4%, 2.21%-87.1%, and 21.8%-72.7%, respectively. The distribution profiles were varied with the coal type and the operation conditions. The Hg in flue gas could be removed by ESP and FGD systems with an average removal efficiency of 51.8%. The calculated average emission factor was  $0.066 \ g/ton$  and much lower than the results obtained ten years ago.

**Key words**: mercury; power plant; mass balance; distribution; removal efficiency; emission factor **DOI**: 10.1016/S1001-0742(10)60584-0

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# Introduction

Mercury has been recognized as one of the priority pollutants in environment since 1970s. Coal combustion is the major anthropogenic emission source of mercury in the world. Pacyna et al. (2006) observed that two thirds ( $1.46 \times$  $10^3$  ton) of the total atmospheric mercury emission (2.19 ×  $10^3$  ton) were originated from coal combustion. Recently, the rapid economic and industrial developments in China have led to the dramatic increase in power demand and coal consumption. It was estimated that the amount of coal consumed in China  $(1.27 \times 10^9 \text{ ton})$  occupied 21% of the world's total coal consumption in 1990 and it has been increased to 25% in 2001, and will be 35% in 2015 (Koukouzas et al., 2006). In 2003, the total power generation capacity has reached 391 GW and among them, that through coal fire shared about 70% (Mukherjee et al., 2008).

Mercury emission from Chinese stationary coal-fired power plants was estimated to be 213.5 ton in 1995 (Wang et al., 1996), 202.4 ton in 1999 (Streets et al., 2005), and

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220 ton in 2000 (Jiang et al., 2005). According to the report of the 25th Session of the Governing Council/Global Ministerial Environment Forum (UNEP, 2009), global atmospheric emissions of mercury from human activities in 2005 were estimated to be approximately 1930 ton (range 1230–2890 ton). However, there are many uncertainties (up to  $\pm$  40%) in the estimate due to the lack of actual measurement data in Chinese sources, such as the mercury concentration and other components in the coal and the technologies both for the production means and mercury pollution control.

In China, most of the coal-fired power plants have been equipped with both the cold-side electrostatic precipitator (ESP) and wet flue gas desulfurization (FGD) for the particles and SO<sub>2</sub> removals respectively. Additionally, the selective catalytic reduction (SCR) system has been added to control nitrogen oxides (NOx). Previous studies indicate that SCR, ESP and FGD can affect the mercury speciation and remove part of the mercury from flue gas. Reviews of EPA's Information Collection Request (ICR) data on mercury capture in boilers and existing air pollution control devices (APCDs) indicate that the mercury removals through cold-side ESP approach to 27%, compared to 4% via hot-side ESP. Both wet and dry FGD systems can remove 80% to 90% of the gaseous mercury (Hg<sup>2+</sup>), and has no effect on the elemental mercury (Hg<sup>0</sup>) removal (Pavlish et al., 2003). Richardson et al. (2002) indicated Hg<sup>0</sup> could be oxidized to Hg<sup>2+</sup> on the surface of SCR catalyst with a high catalytic oxidation efficiency of 80%–90%. The produced Hg<sup>2+</sup> could be absorbed by the wet slurry in FGD system subsequently. However, few researches on the mercury control by existing APCDs and mercury emission factors for Chinese power plants have been reported, which leads to an erroneous estimated value of the mercury emission in China.

In this study, four coal-fired power plants in China were selected to practice onsite mercury measurements, to discuss the mercury removals by existing APCDs and to estimate the fate of mercury from coal-fired power plants. All the gaseous and solid samples, including pulverized coal, bottom ash, fly ash captured in ESP and gypsum in the FGD, were collected at each coal-fired power plant. The Hg in flue gas was trapped with aluminum matrix sorbent and its emissions and removals were estimated. The overall mercury mass balances and mercury emission factors were discussed based on the data in each power plant.

# 1 Methods

# 1.1 Sorbent trap preparation

Active alumina  $(\gamma$ -Al<sub>2</sub>O<sub>3</sub>) loading with metal oxides was prepared as the sorbent following the literature (Mei et al., 2008a, 2008b), which can convert Hg<sup>0</sup> to Hg<sup>2+</sup> by catalytic oxidation and then absorb it. The sorbent was baked in a muffle furnace at 400°C for 2 hr prior to use.

Mercury in flue gas was collected using a sorbent trap with three tubes connected by Teflon sleeves. All the tubes were made by the quartz tube (50 mm in length with an outer diameter of 10 mm and an inner diameter of 7.5 mm) containing approximately 1 g sorbent which was sandwiched and fixed by the quartz cotton. Among them, the tube 1 was designated for primary capture of gaseous Hg. The tube 2 was designated as a backup to check vapor phase  $Hg^0$  breakthrough. For the third tube, a known mass of  $Hg^0$  was spiked to perform the analytical bias test and spiked  $Hg^0$  recovery study. Each sorbent trap was marked with an identified number for tracking.

#### 1.2 Sampling

As shown in Fig. 1, the test gases were sampled from the flue gas stream at an appropriate flow rate (0.3-0.5 L/min) through a heated probe which prevented the condensation occur before the gas entered the sorbent trap(s). A water knockout after the sorbent trap was placed to prevent the water in sample line from entering the flue gas sampler (TH-600, Tianhong, China). The samples were simultaneously taken from both the inlet and outlet of the pollution control devices, e.g., ESP and FGD, as shown in Fig. 2. According to the results of pretest, two hours were enough for the gas phase mercury detection. After the sampling, each tube of the sorbent trap was immediately placed into Hg-free containers for subsequent laboratory detection. The characteristics (such as gas temperature, static pressure and gas velocity) and main components (such as  $SO_2$ ,  $NO_x$  and  $O_2$ ) of the flue gas were determined by the flue gas analyzer (350-XL, Testo, Germany) during the sampling period. Solid samples including the pulverized coal, bottom ash, fly ash captured in ESP and gypsum in the FGD, were taken after half-hour of Hg trapping. The sampling was conducted two or three times for each power plant to obtain the parallel results and reduce uncertainties. The data obtained from tests at the same conditions were averaged to get the final results.

# 1.3 Analyses

The individual tube of the sorbent trap and their respective components were detected separately. Sorbent media, quartz cotton and fly ash deposited on the quartz cotton in each tube were moved into a glass funnel, which covered with a piece of filter paper. Then, they were eluted by 50 mL of 4 mol/L HCl, saturated KCl solution, and the captured mercury was transferred into liquid solution. The mercury concentrations were measured on the cold





**Fig. 2** Simultaneous power plant sampling locations. ESP: electrostatic precipiator; FGD: flue gas desulfurization.

vapor atomic absorption spectrometry (CVAAS) (SG-921, Jiangfen, China). The amount of Hg contents in tube 1 and tube 2 reflected the total mercury concentration in the flue gas. From the results detected in tube 3 spiked with known mass of Hg<sup>0</sup>, the recovery and performance of the combined sampling and analytical approach could be verified. The results detected in tube 1 and tube 2 are reliable only when the recovery of Hg<sup>0</sup> in tube 3 is between 90% and 110%. Coal samples were digested in a V<sub>2</sub>O<sub>5</sub>-HNO<sub>3</sub>-H<sub>2</sub>SO<sub>4</sub> solution firstly and the Hg concentrations of the digestive solutions were determined by a CVAAS (SG-921, Jiangfen, China). The solid samples taken from the power plants were measured by a Zeeman Mercury Spectrometer (RA-915<sup>+</sup>, Lumex, Russia). All the instruments were calibrated before the samples analyses.

# 2 Plant and coal data

#### 2.1 Power plants data

The results about the four power plants tested in this study are summarized in Table 1. Three of the four power plants have pulverized coal power generation units with different capacities and the fourth has an advanced 758 MW supercritical boiler. All the four power plants are equipped with tangential burners, cold-side ESP and wet FGD systems where the watered limestone slurry is used to remove sulfur oxides and gypsum is produced as the byproduct.

#### 2.2 Coal analyses

Recently, the annual coal consumption by the electric utilities in China approximates to  $1.6 \times 10^9$  ton. Ren et al. (2006) compiled Hg data of 1413 coal samples and calculated an average Hg content of 0.20 mg/kg. Zhang et al. (1999) reported an average Hg content in Chinese coals of 0.16 mg/kg based on 990 coal samples, which is higher than that of the world average (0.1 mg/kg) and slightly lower than that of the U.S. average (0.17 mg/kg) (Zheng et al., 2007). Wang et al. (1996) evaluated the Hg content in Chinese coals and calculated an average Hg content of 0.22 mg/kg.

Here, the coals burned in first three plants were from the north China and that in the last plant was from the location which has a higher Hg content than other area in China. Proximate and ultimate analyses of the burned coals are shown in Table 2.

# **3** Results and discussion

## 3.1 Mercury mass balances

Mercury mass balances were estimated and the results are shown in Table 3. The operation parameters such as coal/lime feeding rate, bottom ash production rate, fly ash and gypsum generation rate were provided from operational records by power plants.

The mercury mass balances were 91.6%, 77.1%, 118% and 85.8% for the four power plants, respectively. Some uncertainties, which mainly come from the inhomogeneity of the components in the coal/ash/gypsum, slight variations of boiler operating conditions, and the errors of sampling and chemical analysis procedures, will affect the accuracy of the mass balance estimation. If the mass balance closures are between 70% and 130%, the results are acceptable (Yokoyama et al., 2000). All the results of the four power plants were within this acceptable range.

 Table 1
 Test results of the four power plants

Power plant no.	Boiler type	Coal type	Capacity (MW)	Pollution control device
1	Pulverized coal	Bituminous coal	98	Cold-side ESP + wet FGD
2	Supercritical	Bituminous coal	758	Cold-side ESP + wet FGD
3	Pulverized coal	Bituminous coal	100	Cold-side ESP + wet FGD
4	Pulverized coal	Anthracite coal	162	Cold-side ESP + wet FGD

ESP: electrostatic precipitator; FGD: flue gas desulfurization.

Table 2	Proximate and	ultimate a	nalyses o	of the	burned	coals (	drv	air	basis	)
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Component	Coal-1	Coal-2	Coal-3	Coal-4
Volatile (%)	37.3	34.9	33.0	9.86
Ash (%)	31.6	11.6	23.4	36.9
Moisture (%)	8.50	3.43	4.40	1.88
S (%)	0.570	0.540	0.920	2.41
N (%)	0.790	0.860	0.990	1.15
C (%)	55.3	49.4	54.4	61.4
Cl (mg/kg)	628	472	663	940 <b>0</b> °
Hg (mg/kg)	$0.161 \pm 0.029$	$0.0640 \pm 0.0035$	$0.225\pm0.054$	$0.320 \pm 0.091$

	Stream	Plant-1	Plant-2	Plant-3	Plant-4	
Input	Coal (g/hr)	4.48	16.9	8.24	18.1	
Output	Bottom ash (g/hr)	$5.00 \times 10^{-3}$	$2.00 \times 10^{-3}$	$3.00 \times 10^{-3}$	$452 \times 10^{-3}$	
	Fly ash (g/hr)	0.193	0.366	0.721	4.23	
	Gypsum (g/hr)	2.60	0.373	7.18	3.84	
	Stack flue gas (g/hr)	1.31	12.3	1.79	7.02	
Mass balance	(Input/output) × 100 (%)	91.6	77.1	118	85.8	

**Table 3** Mass balances of Hg between input and output streams in the four power plants (n = 3)

Table 4 Relative distribution of Hg and the mean concentrations of Hg in solid/gas samples in the four power plants (n = 3)

Plant No.		Plant 1	Plant 2	Plant 3	Plant 4
Distribution	Bottom ash (%)	0.110	$1.00 \times 10^{-2}$	$4.00 \times 10^{-2}$	2.50
	ESP (%)	4.30	2.17	8.75	23.4
	WFGD (%)	58.0	2.21	87.1	21.2
	Stack (%)	29.2	72.7	21.8	38.8
Concentration	Bottom ash $(\mu g/kg)$	$6.96 \pm 1.16$	$0.420 \pm 0.996$	$3.25 \pm 3.86$	$49.3 \pm 2.62$
	Fly ash (µg/kg)	$30.6 \pm 5.50$	$8.50 \pm 1.31$	$92.2 \pm 28.6$	$68.5 \pm 29.0$
	Gypsum (µg/kg)	$156 \pm 6.46$	$6.22 \pm 1.86$	$1560 \pm 74.3$	$231 \pm 64.9$
	Stack flue gas ( $\mu g/m^3$ )	$2.27\pm0.58$	$4.29 \pm 0.45$	$1.56\pm0.68$	$5.95 \pm 1.80$

Some other previous investigations are in relatively similar ranges of recovery. Zhang et al. (2008) calculated the Hg mass balances of six power plants, which varied from 100.3% to 139.5%. Yokoyama et al. (2000) estimated the mass balances of Hg for combustion of three type coals were between 89% and 138%. The recovery rates of 82% and 99% were referred by Lee et al. (2006).

#### 3.2 Distribution of mercury

During the process of coal stream in power plant, the mercury in coal is distributed into bottom ash, ESP ash (fly ash), FGD reactant (gypsum) and flue gas. The relative distribution of Hg and the Hg concentrations in solid/gas samples in four power plants are summarized in Table 4. These values were the average of the data which varied depending on factors such as the coal type and the operation conditions of plants.

Little mercury was found in the bottom ashes ranged from 0.11% to 2.50%, which is consistent with the results of previous investigation (Yokoyama et al., 2000; Meij et al., 2002; Lee et al., 2006). The mercury concentration in the bottom ash of plant 4 was far higher than those of the other three plants, which is mainly caused by the difference of coal types. The coal burned in plant 4 was anthracite coal with low volatile (< 10%) and those in first three plants were bituminous coals with higher volatile (20%–40%). Anthracite coal is easy to slag and agglomerate during combustion and covered by melted inclusions which would deteriorate its burn out capacity (Du and Zhang, 1994). Therefore, incompletely burning of coal couldn't lead to a thorough emission of mercury.

The relative distribution of Hg in ESP ranged from 2.17% to 23.4%. During coal combustion, the majority of mercury evaporated in the boiler. When temperature decreases, some gaseous mercury can be condensed or adsorbed on the surface of fly ash particles and collected in the ESP (Zhang et al., 2008). For plant 4, about one quarter of Hg (23.4%) is removed through the ESP and more efficient than others. High concentration of Hg in the coal and great production rate of fly ash may be the major

reasons for the diversity.

Some oxidized mercury in flue gas could be absorbed by lime-slurry when mixed in the FGD scrubber tower due to its water-soluble property (Brown et al., 1999). The relative distribution of Hg in FGD for the four power plants ranges from 2.21% to 87.1%, which varies with the coal types, tower types and liquid-to-gas (L/G) ratios. Hg<sup>0</sup> can be converted into Hg<sup>2+</sup> by reacting with chlorine ions in flue gas (Hall et al., 1991). As shown in Table 2, the concentration of Cl<sup>-</sup> in coal-1 is similar to the one in coal-3. However, the distribution of Hg in FGD in plant 1 is much lower than that of plant 3, which indicates that Cl<sup>-</sup> concentration in coal is not the major factor to control the Hg removal efficiency of FGD. In addition, although the highest concentrations of Hg and Cl- are found in coal-4, the distribution of Hg in FGD in plant 4 only reaches half of the average level (42.2%) of four plants. The highest concentrations of SO<sub>2</sub> (2.41%) in coal-4 may be the important reasons. The oxidation of Hg<sup>0</sup> would be restrained by SO<sub>2</sub> in flue gas in the presence of HCl (Ghorishi et al., 2005).

In stacks, the distribution of Hg ranged from 21.8% to 72.7%, and the final emissions to the atmosphere from stacks were ranged from 1.56–5.95  $\mu$ g/m<sup>3</sup>. Compared with other studies, such as Guo et al. (2007) with a range of 13–21  $\mu$ g/m<sup>3</sup> (with 0.33 mg/kg Hg in coal), Otero-Rey et al. (2003) with a range of 19.1–21.8  $\mu$ g/m<sup>3</sup> (with 0.15–0.17 mg/kg Hg in coal), Lee et al. (2006) with a range of 1.03–2.41  $\mu$ g/m<sup>3</sup> (with 0.043–0.078 mg/kg Hg in coal) and Lei et al. (2007) with a range of 0–48.1  $\mu$ g/m<sup>3</sup> (with 0.011–0.28 mg/kg Hg in coal), Hg emissions from the four power plants were relatively low.

# 3.3 Mercury removals by existing air pollutants control devices

Above results showed that existing air pollution control devices (cold-side ESP and wet FGD) in power plants can remove a part of Hg from flue gas. Here, the average mercury removal efficiency of ESP+FGD systems is 51.8%, corresponding with the US EPA results which ranged from 29% to 74% (US EPA, 1999, 2002a, 2002b). Recently, ESP+FGD or fabric filter (FF) +FGD systems have been applied in most of the power plants in China. Considerable numbers of fluidized bed combustion (FBC) boilers, which promote the adsorption of Hg by fly ash remarkably, have been built for power generation. Additionally, technologies for mercury control in flue gas are being researched and developed. Although the coal consumption in power industry increases annually, these APCDs may play an efficient role on the reduction of mercury emissions, which should be considered when total Hg emission from power plants is estimated for China recently.

#### 3.4 Mercury emission factors

Mercury emission factor is a very important parameter to estimate the total mercury emission. Pacyna et al. (2006) estimated the global mercury emission from power plants in 2000 with the factor 0.1-0.3 g/ton and the emission factor of Chinese power plants in 1999 calculated by Streets et al. (2005) was 0.136 g/ton. In this study, the mercury emission factors were calculated as 0.045-0.124 g/ton and with an average of 0.066 g/ton for the four power plants, which was much lower than the results mentioned above. The decline is assigned to the installation of APCDs in Chinese power plants since 2000. In the past decade, the newly-built power plants are equipped with APCDs, at the same time, the pollution control improvements of old plants are being carried out. Therefore, the average mercury emission factor for coal-fired plants was reduced dramatically. Considering the current situation in Chinese power plants, the estimated value of mercury emissions in this study can be correct.

# **4** Conclusions

Studies on Hg emissions and removals by existing pollution control devices in four different coal-fired power plants which are equipped with cold-side ESP and wet FGD systems have been carried out based on the measurement method with aluminum matrix sorbent. A sorbent trap was used to collect vapor phase Hg from flue gas and the Hg on the sorbent trap was measured using CVAAS followed by acid leaching.

The mercury mass balances are 91.6%, 77.1%, 118% and 85.8% for the four power plants respectively. Total Hg concentrations in stack gas are ranged from 1.56–5.95  $\mu$ g/m<sup>3</sup>. The relative distributions of Hg in bottom ash, ESP, FGD and stack range from 0.110% to 2.50%, 2.17% to 23.4%, 2.21% to 87.1% and 21.8% to 72.7%, respectively. The distribution profile varies with the coal type and the operation conditions of the plants. ESP+FGD systems have the ability to remove Hg from flue gas. The average mercury removal efficiency of ESP+FGD systems is 51.8%. The average emission factor of the four power plants in this study is 0.066 g/ton, which is much lower than the results obtained ten years ago. Further study is needed for the Hg speciation in flue gas and its model prediction.

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