

Mercury transformation across particulate control devices in six power plants of China: The co-effect of chlorine and ash composition

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Abstract

This paper presents the results of field measurements on mercury speciation in six power plants of China by applying the Ontario hydro method. During the tests, flue gas was sampled simultaneously before and after particulate control devices (electrostatic precipitator and fabric filter baghouse) along with the pulverized coal, bottom ash and fly ash sampling. The amount of oxidized mercury in gas phase before and after ESP/FF suggests that mercury oxidation after combustion is a kinetically controlled process. The comparison of mercury speciation in different power plant indicates a clear relationship with coal type, especially the chlorine concentration and the basic ash compositions in coal. Both of the factors are analyzed quantitatively in this study. A new parameter *C* (ratio of chlorine in coal to base/acid ratio) has been introduced to evaluate the co-effect of the two factors above on mercury speciation.

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1. Introduction

Mercury emission is a global air pollution problem attracts more and more attentions world-wide. Of all the anthropogenic mercury emissions, coal combustion has been reported as the largest single category [1,2], contributing about one third of total mercury emission. A three-phase research on the national utility boilers has been organized by US EPA with co-operation from DOE, NETL, EPRI and other related universities and institutes. For the purpose of establishing mercury emission regulations, US EPA collected information about existing coal-fired utility boilers, classified American coals into sub-categories of different mercury speciation characteristics, and investigated the emission factors of power generating units to

estimate mercury emission. Based on the results of the investigation, a rule regulating mercury emissions from coal-fired power plants has been announced under the authority of the Clean Air Act since March 15, 2005 [3]. As specified in the rule, the mercury emission from utility boilers will be reduced to a final cap at 15 ton/year in 2018.

Coal is a more important energy source in China than it is in America, and the annual consumption is about 1.95 billion tons in 2004. Wang et al. [4] has estimated that, the average mercury concentration in Chinese coals is 0.22 mg/kg; whilst USGS has also measured 331 coal samples from different Chinese regions, suggesting that the mercury concentration ranges from 0.02 mg/kg to 0.54 mg/kg, averaged at 0.15 mg/kg [5]. Based on these investigation, Wang et al. [4] and Jiang et al. [5] have calculated the total mercury emissions from coal combustion in China as 302.9 tons (in year 1995) and 161.6/219.5 (based on different databases) tons (in 2000), respectively.

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However, due to the lack of investigation on actual mercury emissions from power plants in China, some fundamental data obtained in US, such as the emission factors for various particulate control devices (ESP/FF) and the mercury speciation, were directly cited in the estimations above. As ash contents in Chinese coals for power generation are normally much higher than those in American coals because of the limited use of coal washing in China, the role of ash compositions must be taken into account. To eliminate these ambiguities in Chinese mercury emission research, the best way is to actually measure the mercury emission characteristics from a series of full-scale utility boilers of Chinese power plants. In this study, onsite measurements have been performed to collect the mercury speciation and partition information across various particulate control devices, including electrostatic precipitator (ESP) and fabric filter (FF) baghouse, in 6 Chinese power plants. The coal properties have also been comprehensively analyzed to investigate their effects on mercury conversion. Both the chlorine concentration and the coal ash compositions have been considered as the key factors affecting mercury transformation.

2. Experimental

2.1. Utility boilers

The utility boilers' capacities of the 6 tested power plants range from 50 MW to 600 MW. These boilers are carefully selected so as to reflect the effects of various types of coals, boilers and particulate control devices. All the 6 power plants are located in the electricity-demanding regions in China so that the obtained mercury emission results could be considered as representative. The details of these boilers' configuration are given in Table 1.

To investigate how the coal type affects mercury speciation, two boilers of similar type but consuming different coals are selected (power plant 2 and 3). Likewise, to investigate how the boiler capacity influences mercury emission, two boilers of the same type, similar coals and particulate control devices but in different capacities are selected (power plant 4 and 5). A circulating fluid bed (CFB) boiler has been chosen since this type of boilers is widely used in

China as a major application of clean coal combustion technology.

2.2. Coal analysis

The coals consumed in five of the tested six power plants are typical coals used in China. Both Shenhua and Zhungeer coal are produced in the northern region. The overall production of these two coals exceeded 50 million tons and 13 million tons, respectively, in 2002, more than 1/20 of the national overall annual coal production. Proximate and elemental analyses of the coals in the 6 power plants are presented in Table 2. All the coals are bituminous, but with different chlorine concentration and ash compositions (Table 5).

2.3. Test methods

Ontario hydro method (OHM) [6] has been adopted to sample the elemental, oxidized, and particle-bound mercury in flue gas before and after ESP/FF simultaneously. Pulverized coal samples from primary air tube, fly ash samples from ESP hoppers or FF baghouses, and slag samples from boiler's bottoms were collected at half-hour intervals during the flue gas sampling process. The location of sampling points for each power plant is illustrated in Fig. 1.

To establish the mercury balance of the boiler system, it is essential to collect all the samples in the same time span in order to minimize the effects of random factors, such as coal feed variation and/or boiler load fluctuation. During the sampling process, the flue gas compositions including O₂ and SO₂ concentrations have been continuously recorded as a clue to monitor both the sampling process and boiler operating conditions. For each power plant, the onsite mercury sampling process has been repeated two or three times under the same operating conditions.

The preparation of all the chemical solutions and the execution of the sampling process strictly followed the OHM requirements. The KCl, H₂O₂ and KMnO₄ solutions in sampling train were digested and analyzed according to OHM specification immediately after the sample collection. The particulate matter (PM) collected by fabric filters in probe nozzle were weighted and analyzed by EPA method 7473. Other gas component such as H₂O, O₂, and SO₂ were

Table 1
Information of onsite tested utility boilers

Power plant No.	Location	Boiler type	Fuel type	Power capacity (MW)	PM control device
Power plant 1	Beijing	WG-Babcock tower shape boiler, W-flame, super high pressure, liquid bottom ash	Shenhua #1	220	Cold ESP
Power plant 2	Hebei Province	SG tangential fired, Sub-critical II boiler	Shenhua #2	600	Cold ESP + FGD
Power plant 3	Inner Mongolia	HG-CE tangential fired, sub-critical II boiler	Zhungeer #1	600	Cold ESP
Power plant 4	Inner Mongolia	HG II boiler	Zhungeer #2	50	FF baghouse
Power plant 5	Inner Mongolia	HG tangential fired, super high pressure II boiler	Zhungeer #3	200	FF baghouse
Power plant 6	Jiangsu Province	HG-ALSTOM super high pressure CFB boiler	Blends ^a	135	Cold ESP

^a Blends vary due to diverse fuel supplies.

Table 2
Summary of coal analysis

Coal type	Shenhua #1	Shenhua #2	Zhungeer #1	Zhungeer #2	Zhungeer #3	Blends
<i>Average proximate analysis, air dry basis (%)</i>						
Moisture	9.74	2.63	4.23	4.48	5.13	4.07
Ash	7.45	30.6	35.7	29.9	28.8	28.9
Volatile	30.2	28.5	24.2	26.9	27.5	24.2
Fixed carbon	52.7	38.3	35.9	36.8	38.6	42.9
Heat value (MJ/kg)	27.08	21.12	17.38	19.99	20.15	21.16
<i>Elemental analysis, air dry basis</i>						
Cl (ppm)	152	510	202	277	290	875
Hg (ppm)	0.011	0.091	0.210	0.278	0.223	0.073
S (%)	0.36	0.51	0.52	0.78	0.83	0.73

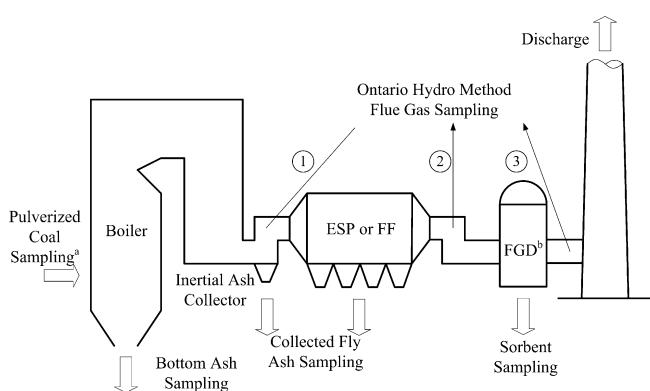


Fig. 1. The onsite sampling positions: (a) except power plant 6 (CFB boiler), all the other power plants in this study have their coals pulverized and (b) only power plant 2 has FGD device, the FGD effect on mercury concentration will not be discussed in this paper.

also analyzed according to related EPA methods, the sampling and analysis method are summarized in Table 3.

3. Results and discussion

3.1. Mercury speciation in flue gas

All the samplings and measurements were taken whilst the boilers were at their full loads. The gas phase elemental,

oxidized mercury and particle-bound mercury concentrations before and after ESP/FF, as well as the SO₂ concentration before ESP/FF, are presented in Table 4. All the data are normalized to dry flue gas in standard conditions with 3% O₂ concentration. Average mass balance for each power plant is given by taking coal mercury as 100%.

mass balance before ESP/FF

$$= \frac{\text{Hg}^T}{\text{Hg}_{\text{coal}}} = \frac{\text{Hg}_{\text{bottom ash}} + \text{Hg}^{2+} + \text{Hg}^0 + \text{Hg}_P}{\text{Hg}_{\text{coal}}} \quad (1)$$

mass balance after ESP/FF

$$= \frac{\text{Hg}^T}{\text{Hg}_{\text{coal}}} = \frac{\text{Hg}_{\text{bottom ash}} + \text{Hg}_{\text{ESP/FF ash}} + \text{Hg}^{2+} + \text{Hg}^0 + \text{Hg}_P}{\text{Hg}_{\text{coal}}} \quad (2)$$

The values presented are the averages of three or two repeated trials under the same conditions along with standard deviations.

In general, the mercury remaining in boiler bottom ash is negligible, less than 1% of all the mercury emission in this study, the detailed data and mass balance will be discussed elsewhere in our further studies; the same conclusion was also reported by Yokoyama et al. [7]. From the testing results listed in Table 4, the following observations can be obtained straightforward.

Table 3
Sampling and analysis methods used in tests

<i>Sampling methods</i>	
Sample and velocity traverses	EPA method 1
Sampling of mercury in flue gas	Ontario hydro method
<i>Analysis methods</i>	
Proximate and heat value analysis	GB ^a 212-91, GB/T 213-1996
Chlorine/sulfur concentration in coal	GB/T ^a 3558-1996/GB/T 214-1996
Mercury concentration analysis of solutions	Ontario hydro method (Standard chemical digestion and CVAAS)
Mercury in coal/PM	EPA Method 7473 Thermal Decomposition, Amalgamation, AAS
Flue gas compositions (O ₂ , H ₂ O)	EPA method 3/4
Fly ash composition	X-ray fluorescence spectrum (XRF)

^a National standard method of China.

Table 4
Mercury speciation concentrations in flue gas before and after ESP/FF

Power plant series no.	Before ESP/FF ($\mu\text{g}/\text{dNm}^3$) In 3% O ₂			Mass balance (%)			After ESP/FF ($\mu\text{g}/\text{dNm}^3$) In 3% O ₂			Mass balance (%)			QA/QC	Other composition SO ₂ (ppm)
	Hg ²⁺	Hg ⁰	Hg _p	Hg ²⁺ /Hg _{coal}	Hg _p	Hg ⁰	Hg ²⁺	Hg ⁰	Hg _p	Hg ²⁺ /Hg _{coal}	Hg _p	Hg ⁰		
1	0.13 (0.04) ^a	1.42 (0.57)	0.02 (0.01)	121.79	0.10 (0.00)	1.08 (0.35)	ND ^b	94.17	Tr ^c	132				
2	0.25 (0.13)	6.30 (0.59)	0.21 (0.15)	79.22	0.94 (0.26)	7.18 (1.75)	ND ^b	101.60	Tr ^c	266				
3	6.27 (0.81)	41.80 (8.46)	1.99 (0.89)	124.64	14.54 (4.24)	17.55 (3.52)	0.01 (0.00)	100.29	Tr ^c	206				
4	14.17 (4.78)	4.91 (3.31)	27.99 (10.91)	95.17	7.43 (3.29)	1.14 (0.42)	2.91 (0.93)	109.92	Tr ^c	406				
5	21.58 (0.92)	18.46 (3.85)	8.65 (4.50)	132.23	29.46 (2.43)	9.55 (0.41)	ND	127.27	Det ^c	575				
6	ND ^b	ND ^b	11.10 (2.49)	116.37	ND ^b	ND ^b	0.17 (0.01)	138.56	Tr ^c	NA ^b				

^a Values in brackets stand for the standard deviation of 2 or 3 identical tests.

^b Not detected/not available.

^c Triple/double measurement.

- Gaseous mercury (Hg²⁺ and Hg⁰) is detected in the range of virtually 0 to 48.07 $\mu\text{g}/\text{Nm}^3$ in flue gas of different power plants.
- Ignoring the mercury in boiler bottom ash, the total mercury concentrations in flue gas (i.e., gaseous and particle-bound mercury) before the ESP/FF depend on the mercury concentration in the coal, i.e., higher mercury concentration in coal, more mercury in the flue gas.
- The fly ash adsorbing ability determines the mercury partitioning in gas phase and particle-bound phase. The amount of gaseous mercury (Hg⁰ and Hg²⁺) adsorbed by fly ash depends on the characteristics of fly ash, especially LOI (loss on ignition), which is shown in Table 5. In this study, the higher LOI, the more mercury was adsorbed by fly ash.
- Since more than 99% of particle-bound mercury is removed by ESP or FF baghouse, the amount of Hg²⁺ and Hg⁰ in gaseous form determines the final total mercury emitted to atmosphere from stacks. However, power plant 4 is an exception. Due to its poor FF performance, considerable amount of particle-bound mercury was discharged with flue gas.
- Mercury speciation in gas phase varies in different situations, because Hg²⁺ could be removed by WFGD, the factors affecting mercury conversion from elemental to oxidized are important and are analyzed in later sections.

The high mercury removal efficiency (nearly 100%) in CFB boiler of power plant 6 is interesting. Gas phase mercury is hardly detected in flue gas both before and after ESP, while the mercury concentration as high as 0.48–0.78 ppm is detected in the particulate matter collected from the flue gas, and 0.55–1.25 ppm is detected in hopper ash from the first two ESP electrical fields (0.073 ppm in coal in comparison). Three main causes may attribute to this high particle-bound mercury concentration, i.e., high chlorine concentration in coal (875 ppm), high LOI (10.8%) in fly ash, low combustion temperature (900 °C) and strong interaction between flue gas and particulate matter in fluidized bed and cyclone separator.

3.2. Mercury oxidation across ESPIFF

Although gas phase mercury is barely removed across the ESP/FF by fly ash with low LOI, it does transform continuously across the ducts and devices. The proportions of oxidized mercury in gas phase mercury are increased across either ESP or FF as shown in Fig. 2, where Hg_{GT} means mercury in gas phase, and the figures beside the symbols identify the measured power plant no.

Equilibrium calculations indicate that Hg⁰ is almost completely oxidized in the form of Hg²⁺X and Hg_p when cooling to 400 °C [8,9], however, the measurements in this study show that only 10% to 87% mercury was oxidized at the outlet of ESP/FF, where the flue gas temperature is about 160 °C indicating that mercury oxidation in flue gas is kinetically controlled, and mercury is oxidized con-

Table 5
Summary of fly ash composition collected in flue gas before ESP/FF in 6 power plants

Element in the form of oxide (%)		Power plant series no.					
		1	2	3	4	5	6
		Shenhua #1	Shenhua #2	Zhungeer #1	Zhungeer #2	Zhungeer #3	Blends
Basic oxides	Fe ₂ O ₃	11.08	6.89	2.76	2.35	2.29	2.44
	CaO	22.85	13.26	3.59	2.89	2.17	3.06
	MgO	1.97	1.35	0.72	0.31	0.28	0.53
	Na ₂ O	1.18	0.95	0.16	ND ^a	0.12	0.15
	K ₂ O	1.14	1.15	0.64	0.42	0.33	0.75
Acid oxides	SiO ₂	41.76	44.12	48.46	45.39	49.53	42.83
	Al ₂ O ₃	14.49	24.93	39.97	42.38	42.51	29.64
	TiO ₂	0.79	1.4	2.03	1.34	1.95	1.18
Chlorine	Cl	0.03	0.02	0.02	ND	ND	0.03
LOI of fly ash		2.59	1.69	0.90	3.14	1.15	10.8
Base/acid ratio		0.671	0.336	0.087	0.067	0.055	0.094

^a ND: Not detected due to low concentration.

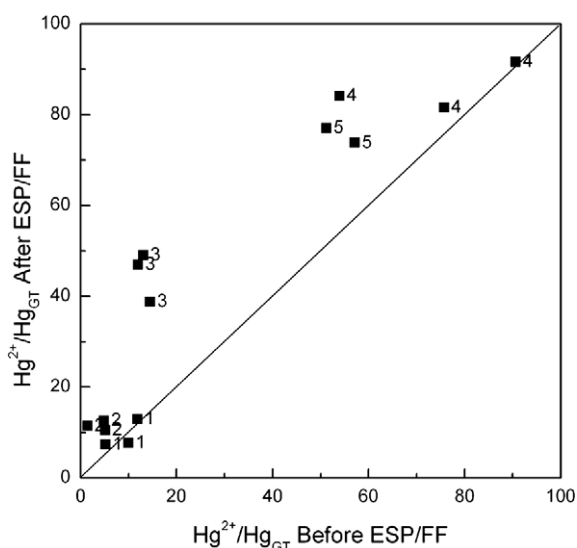


Fig. 2. Mercury oxidation across ESP/FF in 5 power plants.

tinuously in the residence time across ESP/FF. The typical designed flue gas velocity in ESP is 1 m/s, so the total residence time is about 15 s in a 4-stage ESP. The particulate control devices accommodate the reactions of mercury with space and proper temperature. The oxidized mercury proportion in gas phase has been increased to different degrees, because of different coal types, which will be discussed in later sections.

More efficient oxidations have been achieved in the FF baghouse in power plant 4 and 5, probably due to the extended contact time and enhanced interaction between mercury and oxidants throughout the dust cake covering the fabric surface.

3.3. Effect of chlorine in coals

Mercury is oxidized during the flue gas cooling down process, mainly by chlorine, hydrogen chloride, oxygen

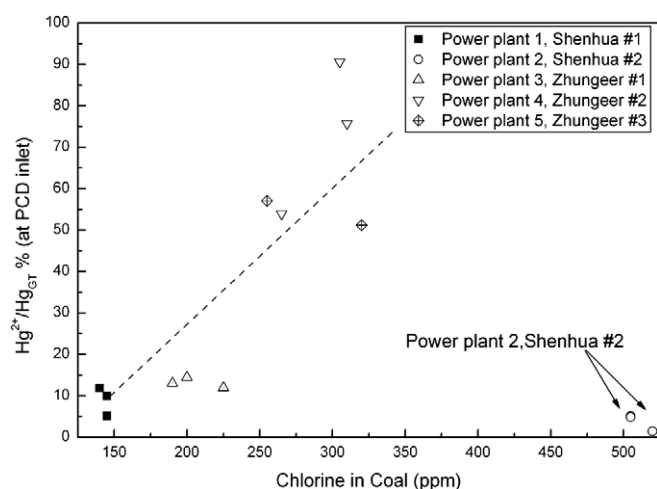


Fig. 3. Mercury oxidation by chlorine in flue gas (at ESP/FF inlet).

and other oxidizing components. In typical flue gas, the only thermodynamically stable species of mercury is elemental when the temperature is higher than 750 °C; Hg⁰ (g) and minor proportions of HgO (g) are predicted to coexist at lower temperature; HgCl₂ (g) is the dominant species in a chlorine-bearing flue gas at <430 °C [9]. The effects of chlorine in flue gas have been reported extensively both in thermodynamic and experimental studies.

In our onsite test results, the proportions of oxidized mercury in gas phase at the inlet of ESP/FF increase with the chlorine concentration in coal as shown in Fig. 3. Temperature may be another influencing factor, however, since the ESP/FF inlets of 6 power plants are at similar temperatures ranging from 139 °C to 166 °C, its influence on mercury speciation cannot be clearly identified and thus is not covered in this paper. Oxidized mercury in total gaseous mercury increases from about 10% to 60% as the chlorine content in coal increases from 150 ppm to 350 ppm. This result is similar to EPA's 1999 ICR data [10], indicating that the mercury speciation for Chinese coals has the

similar trends with the chlorine in coal as that of US coals. However, mercury oxidation of power plant 2 did not fit into this trend, shown as outstanding points in Fig. 2. This phenomenon is discussed in the following section.

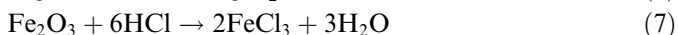
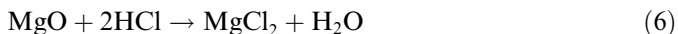
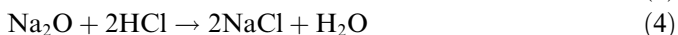
3.4. Effect of coal ash composition

As discussed above, chlorine has a strong effect on mercury oxidation. However, chlorine is not the only factor affecting mercury oxidation. Fujiwara et al. [11] has reported that mercury oxidations are not proportional to coal–Cl level, which is consistent with our onsite test findings. The oxidized mercury percentage is unexpected low with a high coal–chlorine content for Shenhua coal #2 (power plant 2). Considering the same boiler capacity, similar boiler type and combustion means in power plant 2 and 3, a possible explanation is the coal type difference. In this case, coal ash compositions are believed as the key.

The effect of fly ash on mercury oxidation is studied by many researchers, focusing on the catalytic effect of maghemite contents [12,13], however, the effect of basic oxides including CaO, K₂O and Na₂O etc. is usually neglected. As a substantial existence in coal combustion, basic ash compositions should be taken into account as it reacts with acid gas, such as chlorine. Shemwell et al. [14] have conducted a detailed laboratory-scale study on the capture of HCl gas by dry-injection of calcium-based sorbents, suggesting the same composition in coal ash could have the same effect. Lee et al. [15] also reported the mercury oxidation rate reduction due to gas-phase HCl removal by CaO addition in simulated fly ash.

High basic oxide composition is one of the most outstanding features of Shenhua coals, its ash content consists of 15–40% CaO and 5–20% Fe₂O₃[16]. These oxides may compete against mercury to react with HCl and Cl₂, thus inhibit mercury oxidation.

There are several possible reactions between basic oxide and HCl:



To verify this probability, X-ray fluorescence spectrum (XRF) analysis has been adopted to analyze the fly ash collected by the fabric filters of OHM before ESP/FF. The ash compositions are shown in the form of metal oxides in Table 5. It is obvious that the Shenhua coal #1 and #2 consist of much more CaO and Fe₂O₃ than Zhungeer coal #1, #2 and #3.

Assuming that each basic oxide content has the similar capability to react with HCl and Cl₂, the factor BAR (base/acid ratio) is introduced as

$$\text{BAR} = \frac{\text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO} + \text{Na}_2\text{O} + \text{K}_2\text{O}}{\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{TiO}_2} \quad (8)$$

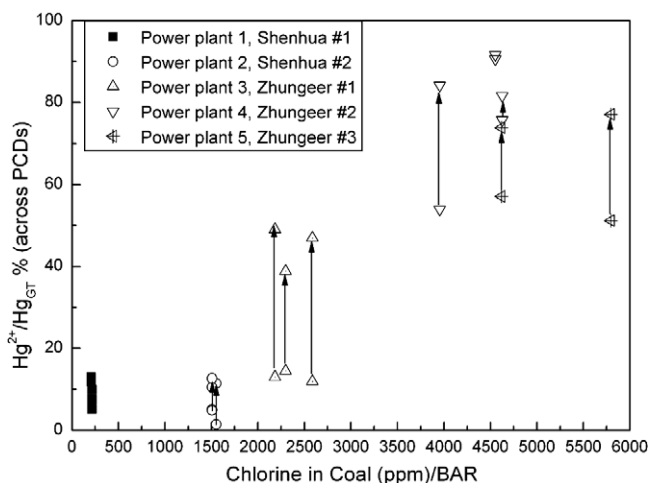


Fig. 4. Mercury oxidation in the function of C , considering both chlorine in coal and basic ash compositions (at ESP/FF inlet).

Defining C as the quotient of chlorine in coal and BAR in coal ash, showing the co-effect of chlorine concentration and basic ash compositions on mercury oxidation.

$$C = \frac{\text{Cl concentration in coal}}{\text{BAR}} \quad (9)$$

Fig. 4 illustrates the trend of mercury oxidized as a function of C , showing better correlation than Fig. 3. In this way, power plant 1 and 2 burning Shenhua coals with low C value give very low Hg²⁺ percentage in gas phase mercury, the mercury oxidation is not obvious in ESP either; on the other hand, the Zhungeer coals burnt in power plant 3, 4 and 5 consist of high acid oxide compositions [17] with high C value show different trend of mercury oxidation. The oxidized mercury proportions in gaseous mercury increase from 15% to 50% for Zhungeer coal #1 (power plant 3), and from 50% to 80–90% for Zhungeer coals #2 and #3 (power plant 4 and 5).

It should be noticed that the ash compositions interpreted by XRF are in oxide form, not the real form of their existence. The parameter C is only attempted in order to comprehend the effect of coal ash composition. More detailed analysis and study is needed in the future.

3.5. Other factors affecting mercury oxidation

Further more, the comparisons of power plant 1 and 5, power plant 2 and 3 (outlet of ESP/FF) suggest that the mercury speciation may be very different even in the similar boilers (and particulate control devices) due to the diversity of coal type. On the other hand, the capacity difference of boilers burning the same coal may not affect the gaseous mercury speciation. For power plant 4 and 5, with capacities of 50 MW and 200 MW, respectively, the mercury speciation is very similar. This seems to suggest that the coal characteristics, rather than the equipment and operation diversities, are the dominant factors affecting mercury speciation. Therefore, it is necessary to pay much more

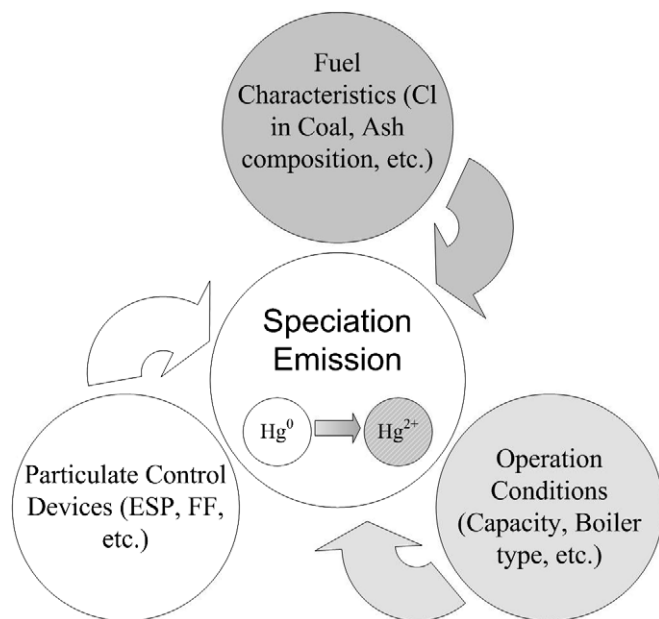


Fig. 5. Schematic of factors affecting mercury speciation in this study.

attention on the coal subcategories when estimating mercury emission and speciation. As discussed in the former section, the residence time is also important, longer residence time in the ducts and ESP/FF will also enhance the mercury oxidation.

A schematic is shown in Fig. 5 to illustrate the factors influencing mercury speciation in coal-fired power plants. The importance order is fuel characteristics, and then operation conditions and particulate control devices.

4. Conclusions

Mercury simultaneous samplings before and after ESP/FF via Ontario hydro method are achieved in 6 power plants of China to investigate the mercury speciation across ESP/FF; coal, bottom ash and fly ash are analyzed to find out the factors influencing mercury speciation transformation. The main conclusions are as following:

1. Continuous oxidation of mercury is observed across the ESP/FF, which suggests the reaction between mercury and oxidants in flue gas is kinetically controlled.
2. Coal characteristics, i.e., chlorine concentration and ash composition in coal, play more important roles in determining gas phase mercury speciation than boiler capacity and particulate control device type do.
3. Chlorine in coal is the major oxidant for mercury oxidation in flue gas, similar to the conclusions from US coals.
4. Ash compositions, especially the basic ash compositions, are also believed to be the key factor affecting mercury oxidation. Basic ash might react with chlorine to compete against Hg. The factor C (quotient of Chlorine in coal over BAR) could be an index of mercury oxidation.

The mercury speciation in all 6 power plants showed a clear correlation with this parameter.

From this onsite research, it has been recognized that Chinese coals are diverse in mercury concentration, chlorine concentration and ash compositions due to their origins and geological history. A more comprehensive mercury investigation on coal classification and power plant emission is needed to obtain more detailed information before estimating the overall mercury emission from Chinese power plants, and determining the reasonable and realizable mercury control strategies for China.

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