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Mass Balance and Isotope Characteristics of Mercury in Two Coal-fired Power Plants in Guizhou, China

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Abstract. Mercury distribution and stable isotope composition in solid samples of two coal-fired power plants in Guizhou province were determined. Results shown electrostatic precipitator (ESP) has mercury removal efficiency between 29.53% to 58.41%, and wet flue gas desulfurization (WFGD) between 12.29% to 58.60%, mercury removal efficiency of ESP and WFGD mainly depends on the coal properties. Most mercury (70% to 88%) in coal was captured by the combination of ESP+WFGD. Mercury in fly ash and gypsum were much heavier in isotope composition compared to the coal, hints mercury escaped into atmosphere was enriched in lighter mercury isotopes.

Introduction

Mercury (Hg) is a global pollutant due to the long range transportations in the atmosphere. Coal combustion is regarded as the largest anthropogenic Hg source to atmosphere in the world, accounting for 45.6% of total anthropogenic emissions in 2005 [1]. China burns large quantities of coal each year, and emits 202 tons of Hg from this source category in 1999, which equivalent to 38% of China's total emissions [2]. About one half of the coal consumed in China is by the coal-fired power plants (CFPPs), therefore, control Hg from CFPPs is critical to China in order to protect the environment.

Most Chinese CFPPs has installed air pollution control devices (APCD) with different configuration to control particulate matter (PM) and SO_2, only a few installed selective catalytic reduction (SCR) or selective non-catalytic reduction (SNCR) to control NOx. For PM controls, electrostatic precipitator (ESP) dominated in over 96% installed capacity, and the other 3% is fabric filter (FF) [3]. While for SO_2, 92.3% is wet scrubber systems, the other is dry (4.05%), semi-dry (2.62%) and CFBC (1.01%) systems [4]. There is a synergistic effect of mercury removal by the conventional pollutant controls, for example, installation of flue gas desulfurization (FGD) during 2005 to 2008 had resulted in 26.2 tons of Hg emission reduction national wide [3].

Guizhou province, ranks No.1 in coal resource in southern China, constructed 15 large CFPPs with a total capacity of about 16000 MW, making it's an important energy exporter in China. All CFPPs in Guizhou has installed ESP for PM control, 14 equipped with limestone wet scrubber system, only one with CFBC for SO_2 control. In order to investigate the fate of Hg in the modern CFPPs in Guizhou, two power plants were investigated.

Sample collection and analysis

Two CFPPs in southwest Guizhou, AS and PX, were studied. AS has four 300 MW units and PX has five 200 MW units, all are PC boilers and equipped with ESP and limestone wet FGD. The coal burned for two CFPPs are different, i.e., anthracite for AS and bituminous for PX, the basic physico-chemical properties of coal from the two power plants is listed in Table 1.

Solid samples from each unit of AS and PX CFPPs, including coal, lime stone, bottom ash, fly ash, FGD slurry, were collected, every solid sample was a mixture of three subsamples taken during a 8-hour period. Total Hg were determined by CV-AAS technique (Lumex RA915+ mercury analyzer with PYRO-915 attachment, Lumex Ltd., Russia), standard samples of coal and fly ash (NIST 1630a and 1633b) were used to control the QA/QC. Hg stable isotope was measured by Multicollector inductively coupled plasma-mass spectrometer (Nu-Plasma MC-ICP/MS, Nu Instruments, Great Britain).

Table 1 Proximate and ultimate analysis of coal samples

Power plant	Proximate analysis					Ultimate analysis				
	M*ad* %	A*ad* %	V*ad* %	FC*ad* %	Q MJ/kg	C*ad* %	H*ad* %	N*ad* %	O*ad* %	S*ad* %
AS CFPP	2.76	40.45	9.79	46.99	19.10	50.36	2.03	0.90	1.19	2.30
PX CFPP	1.34	34.32	19.80	44.55	22.56	55.74	3.38	0.95	3.50	0.78

Hg distribution and mass balance in the power plant

Hg content in solid samples and the flow rate of solid material are listed in Table 2, and Hg input and output flux is calculated therefore and shown in Table 2 and Fig. 1. At these two CFPPs, Hg input from the coal was overwhelming (>99.8%) compare to that from limestone (<0.2%). Most Hg was evaporated during the coal combustion, with only 0.22%-0.23% retained in bottom ash. While, Hg removed by ESP and WFGD was obviously different at AS and PX, AS was dominated by ESP (58.41% Hg remove efficiency), and PX by WFGD (58.60% Hg remove efficiency), the reason maybe is the different properties of coal burned at these two power plant (Table 1). The totally synergetic Hg remove efficiency by the combination of ESP and WFGD at AS and PX was 70% and 88%, respectively, this means, only a small portion Hg (<30%) was escaped from CFPPs through the stack.

Table 2 Hg content in the solid samples and Hg input/output ratios in AS and PX CFPPs

CFPPs	Solid samples		Hg content (ng/g)	Solid material input /output flow (t/h)	Hg flux (g/h)	Hg input/output percentage (%)
AS CFPP (300 MW Unit)	Input	Coal	116	125	14.50	99.86
		Limestone	3	7	0.02	0.14
		Subtotal			**14.52**	**100**
	Output	Bottom ash	4	8	0.03	0.22
		Fly ash	242	35	8.47	58.41
		Gypsum	132	13.5	1.78	12.29
		Subtotal			**10.28**	**70.82**
PX CFPP (200 MW Unit)	Input	Coal	72	108	7.78	99.94
		Limestone	1	4.74	0.005	0.06
		Subtotal			**7.78**	**100**
	Output	Bottom ash	5	3.57	0.02	0.23
		Fly ash	71.5	32.11	2.30	29.53
		Gypsum	635	7.8	4.56	58.60
		Subtotal			**6.87**	**88.35**

Hg isotope composition in coal and coal combustion product

Results of Hg isotope composition in coal and coal combustion product is in Table 3. δ^{202}Hg in two CFPPs coals is similar (-1.44‰ vs -1.38‰), and identity to results of Guiyang's coal (i.e., -1.22‰ ~-1.37‰) [5]. While coal from PX has slightly mass independent fractionation (MIF), namely Δ^{199} Hg and Δ^{202}Hg reached up to 0.31‰ and 0.24‰, respectively, this result demonstrate syngenetic (depositional) sources of Hg in coal [6]. At both power plants, Hg in the fly ash and bottom ash are obviously heavier in δ^{202}Hg compared to coal as shown in Table 3 and Fig.2, this means more Hg featured with lighter Hg isotope is discharged from the stack into ambient air.

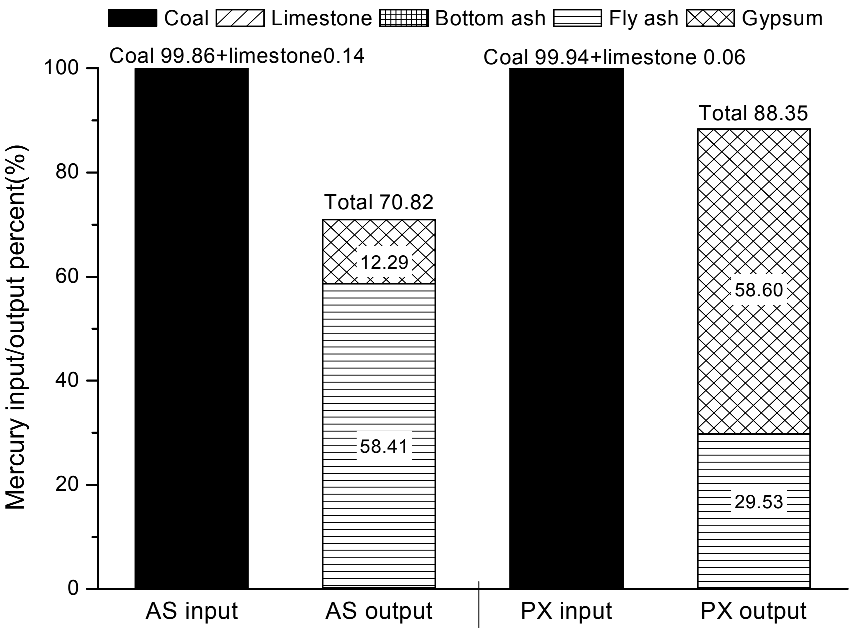

Fig.1. Mercury input and output percentage in AS and PX power plants

Table 3 Hg concentration and isotope ratios of solid samples from AS and PX CFPPs

Sample type	Hg content (ng/g)	δ199 (‰)	δ200 (‰)	δ201 (‰)	δ202 (‰)	Δ199 (‰)	Δ200 (‰)	Δ201 (‰)
AS Coal	116	-0.52	-0.80	-1.04	-1.38	-0.18	-0.11	-0.01
AS fly ash	242	-0.26	-0.52	-0.70	-0.95	-0.02	-0.04	0.01
AS gypsum	132	-0.13	-0.39	-0.78	-0.94	0.11	0.08	-0.08
PX coal	72	-0.05	-0.62	-0.85	-1.44	0.31	0.11	0.24
PX flyash	72	-0.36	-0.03	-0.78	-1.26	-0.04	0.60	0.17
PX gypsum	635	-0.37	-0.46	-0.89	-1.02	-0.11	0.05	-0.12

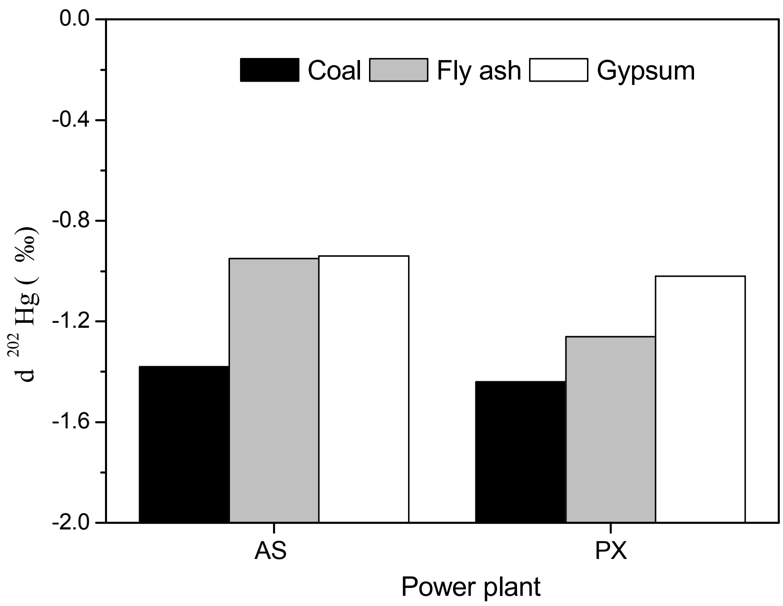

Fig.2 δ^{202}Hg ratios of coal, fly ash and gypsum samples from AX and PX CFPPs

Conclusions

(1) Both ESP and WFGD has synergistic effect of mercury removal in AS and PX CFPPs, while AS dominated by ESP and PX by WFGD, the total Hg removal by the combination of ESP+WFGD is 70% and 88% at AS and PX, respectively.

(2) Hg captured by ESP and WFGD is relative heavier than the coal in terms of Hg isotope ratios at both CFPPs, this hints Hg escaped from the stack should be much lighter than the coal.

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