



Full Length Article

Emission of mercury from six low calorific value coal-fired power plants

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ABSTRACT

The feed fuel, bottom ash, fly ash, limestone and gypsum have been sampled in six low calorific value coal-fired power plants in Shanxi province, China. The emission of mercury (Hg) from the six power plants was conducted. The results showed that about 98% of Hg in feed fuel can be released out in the form of gaseous Hg. Electrostatic precipitator (ESP) and fabric filter (FF) used in the six power plants present higher Hg removal efficiencies. About 58.83–88.00% of Hg is highly enriched in fly ash removal by ESP and FF. Furthermore, less than 12% of Hg is transferred into the gypsum. The average Hg emission factor for low calorific value coal-fired power plants in this study is 52.55 $\mu\text{g}/\text{kW}\cdot\text{h}$, which is significantly higher than that of coal-fired power plants due to the higher Hg content and the lower net calorific value in feed fuel.

1. Introduction

Large amounts of low calorific value (LCV) coals, including coal gangue, coal slime and parts of middlings, are produced from coal mine and coal preparation plant in China each year [1]. Large quantities of LCV coals are dumped and occupied a tremendous amount of lands, potentially causing severe environmental problems by polluting the air, water and soil [2–5]. In recent years, LCV coals are gradually utilized as raw fuel for mine-mouth power generation via co-firing because there are some calorific values in LCV coals. It is considered as a feasible way to reduce the environment pollution as well as bring economic and social benefits [1,6]. According to Annual Report on Comprehensive Utilization of Resources of China (2014), China's installed capacity of LCV coal-fired power plants have reached 30 million kW in 2013. Moreover, the installed capacity of LCV coal-fired power plants is still increasing on the basis of the 13th Five-Year Plan of China, especially in Shanxi province.

It is well known that Hg is an important global air pollutant in the environment because of its volatility, persistence, and bioaccumulation. Coal combustion has been recognized as the major anthropogenic emission source of Hg and power plants are considered to be one main source in most countries [7,8]. As a result, research on the emission of mercury from coal-firing power plants has gained considerable attention in recent years [9,10]. It is reported that the emission behavior of Hg is influenced by the fuel composition, combustion conditions, fly ash properties and air pollutant-control technologies [11–16]. The Hg emissions from coal-fired power plants in China are increasing by 5.9% annually and reaching to 67.97 t, 100.1 t, 172 t in 1999, 2003, 2009,

respectively [17–19].

The LCV coals have higher contents of mineral and less content of organic material than normal coal [20]. Especially, they usually have more content of Hg than the original raw coal [21]. Therefore, the emission behavior of Hg during LCV coals combustion may be significantly different from that of normal coal. Although the LCV coal-fired power plants is rapidly developing, studies on the Hg emission of LCV coal fired power plants are limited and the emission factor of Hg from LCV coal-fired power plants is still unknown thus far. There is only a report about Hg release from a coal gangue-fired power plant and it showed that Hg were highly enriched in fly ash and might be emitted into the environment via the gas phase [20]. However, some basis data about the Hg partitioning behavior and emission factor from LCV coal-fired power plants in various systems are still not clear. Meanwhile, the Hg removal efficiencies of existing pollution control devices in LCV coal-fired power plants are still unknown. Consequently, the investigation of the Hg emissions from LCV coal-fired power plants is essential.

For this purpose, this study focuses on the Hg emission from six LCV coal-fired power plants in Shanxi province, China. The objectives of this study are (1) to provide further knowledge on the Hg partitioning behavior during LCV coal combustion; (2) to estimate the Hg emission factor from LCV coal-fired power plants; (3) to evaluate the Hg removal efficiencies of the typical pollution control devices used in LCV coal-fired power plants.

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Table 1

The configurations of the six LCV coal-fired power plants.

Power plant	Location	Boiler type	Feed fuels	Capacity (MW)	Pollution Control Device
DY	Lvliang	CFB	gangue, middlings	12	ESP + FF + WFGD
YW	Chengzhi	CFB	gangue, raw coal	135	LID + FF
WP	Datong	CFB	gangue, middlings, slime	200	FF + WFGD + SCR
YH	Shuozhou	CFB	gangue, middlings	50	FF + WFGD
TD	Datong	CFB	gangue, middlings	330	LID + ESP + FF
QX	Chengzhi	CFB	gangue, middlings, slime	10	LID + FF

Table 2

Proximate and Ultimate Analyses of feed fuels (wt%).

Samples	Proximate analysis, ad			Ultimate analysis, ad					Q _{net,ad} (MJ/kg)	Hg(ng/g)
	M	A	V	C	H	N	S	O ^a		
DY	1.06	51.44	18.12	35.47	2.40	0.57	3.66	5.40	13.63	749.00
YW	0.60	50.08	10.94	41.40	2.25	0.83	0.25	4.59	15.50	363.00
WP	0.82	53.98	17.52	33.06	2.26	0.56	0.32	9.00	12.00	283.25
YH	1.18	41.80	23.66	42.30	3.03	0.74	2.03	8.92	16.23	646.88
TD	0.77	49.20	18.41	37.80	2.39	0.64	0.31	9.09	13.77	269.25
QX	0.71	45.18	14.73	44.26	2.50	0.64	2.33	4.38	16.82	313.50

ad: air dried basis; M: Moisture; A: ash; V: volatile matter; Q_{net,ad}: Net calorific value; a: by difference.

2. Materials and methods

2.1. Plant description

Shanxi province is the main coal production district in China and some typical LCV coal-fired power plants are located here. The installed capacity of LCV coal-fired power plants reached 7.43 million kW in 2014 (account for about 1/4 of LCV coal-fired power plants in China), and approximately 300 million tons of LCV coal were used in LCV coal-fired power plants in Shanxi province.

The detailed configurations of the six typical LCV coal-fired power plants, which are located in four main coal resources areas in Shanxi province, are given in Table 1. They are Wangping (WP) and Datong Tongda (TD) in Datong city, Qingxin (QX) and Yuwu (YW) in Changzhi city, Dongyi (DY) in Lvliang city and Yonghao (YH) in Shuozhou city. The feed fuel for them is mainly composed of typical LCV coals such as coal gangue, slime, middlings or raw coal. All the tested units in plants are circulating fluid bed (CFB), which are the most typical boilers used in LCV coal combustion technology. The installed capacities of plants range from 10 MW to 330 MW, which almost cover all installed capacity of LCV coal-fired power plants. Typical particulate control devices of ESP or FF are used to capture the fly ash. Limestone injection desulfurization (LID) and wet flue gas desulfurization (WFGD) are applied for SO₂ removal. NO_x-control device of selective catalytic reduction (SCR) is only applied in WP power plant. Overall, the six plants tested in this study are typical in LCV coal-fired power plants in China.

2.2. Samples collection

About 20 kg feed fuel samples for each power plant were obtained from conveying duct connected to the boilers before combustion. About 3 kg bottom ash (dropped below boiler) and fly ash (sequestered by ESP or FF) were sampled shortly after the feed fuel was combusted. About 8 kg limestone and gypsum were collected from the desulfurization system simultaneously during the operation of the boiler. All samples were collected three times in 3 h for each power plant to obtain parallel results and thus to reduce uncertainties. Duplicated samples were mixed to obtain a representative sample, which were pulverized to finer than 100 mesh and air-dried prior to analysis.

2.3. Sample digestion and Hg analysis

The samples were digested in microwave digestion system. On the basis of the method described by literature [21], 0.1 g of each sample was digested by a mixture of 6 ml of HNO₃, 2 ml of HCl and 2 ml of HF in a microwave oven. The liquids obtained from the digestions were solid-free and therefore suitable for Hg analysis using atomic fluorescence spectroscopy (AFS). The detection limit of AFS for Hg in solution is 0.01 ng·mL⁻¹ and the analysis uncertainty of obtained Hg content is less than 3%.

3. Results and discussion

3.1. Properties of feed fuel

Proximate and ultimate analyses of the feed fuel are given in Table 2. It can be seen that all the feed fuels from six plants have a remarkably high ash yield in the range of 41.80–53.98%. The net calorific values of the feed fuels are in the range of 12.00–16.82 MJ/kg, meeting the requirements of LCV coal-fired power plants in Shanxi province (5.02–17.57 MJ/kg) [22]. Total Hg concentrations of feed fuel are in a range of 269.25–749.00 ng/g. Compared with the average Hg concentrations of normal coal in China (typical range of 100–300 ng/g) [23], the Hg concentrations of feed fuel in the present study is overall higher. Also, it shows a broad variation, which is possibly attributed to the different origins of the feed fuel [20].

3.2. Hg partitioning behavior in the byproducts of LCV coal-fired power plants

The concentration of Hg in the byproducts of LCV coal-fired power plants, including bottom ash, fly ash, limestone and gypsum, are shown in Table 3. The relative enrichment factor (RE) is introduced to describe the Hg partitioning behavior in the byproducts and thus to easily compare the levels of Hg enrichment in the byproducts with the feed fuel, which can be calculated according to Eq. (1) [15,20,24]. The result is presented in Fig. 1.

$$RE = \frac{C_x \times A_c}{C_c} \times 100 \quad (1)$$

where C_x is the Hg concentration in bottom ash or fly ash; C_c is the Hg

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