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Migration and emission of mercury from circulating fluidized bed boilers co-firing petroleum coke and coal

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ABSTRACT

The migration and emission of mercury (Hg) were studied for three 410 t/h circulating fluidized bed (CFB) boilers co-firing petroleum coke and coal. Both the Ontario Hydro Method (OHM) and US Environmental Protection Agency (EPA) Method 30B were employed to sample gas phase emissions of mercury from the flue gas, and to compare the agreement for these different measurement methods in industrial application. Concurrent with flue gas sampling, solid and liquid samples including fuel, bottom ash, fly ash and gypsum, wastewater, etc., were also collected to determine the total mass balance and map the mercury migration from the power plant. The results showed that the mass balance rates ranged from 83.9% to 122.7%, which can be considered to be both acceptable and reliable. The vast majority of mercury emitted was distributed in the fly ash and stack gas, accounting for 61.36–67.71% and 22.22–33.35%, respectively. The total Hg concentration measured by OHM is comparable with that determined by EPA Method 30B; however, EPA Method 30B possesses advantages in terms of flexibility. The fabric filter (FF) has better Hg^0 and Hg^{2+} removal efficiencies than the electrostatic precipitator (ESP). Because the Hg contained in the liquid waste streams greatly exceeded Chinese regulations, the main emphasis of future work should be focused on wastewater treatment. The mercury emission factors in this study are in the range of 0.69 g/TJ-0.80 g/TJ, which provides basic data for such CFB power plants in China. The CFB boilers equipped with ESP + WFGD or FF + WFGD appear to have the potential to significantly reduce Hg emission to the atmosphere.

1. Introduction

Mercury (Hg) and its compounds from anthropogenic sources have raised environmental concerns because of their potential to cause persistent health damage, biologically accumulate and to demonstrate extensive mobility [1]. Presently, coal-fired power plants are considered to be the main anthropogenic source for Hg emissions into the atmosphere [2,3]. It was reported that in 2010, about 24% of total global anthropogenic mercury emission was from coal-fired power plants [4]. To cope with the serious Hg pollution, the "Minamata Convention," an international, legally-binding treaty to prevent Hg emissions and release was signed by eighty-six countries including China in October 2013 [5]. The Chinese government also established the latest emission standard for air pollutants from thermal power plants [6], which requires emission values of Hg be limited to 30 $\mu g/m^3$ or less.

The Hg in the flue gas mainly occurs in three forms: gaseous elemental mercury (Hg^0), gaseous oxidized mercury (Hg^{2+}) and particulate-bound mercury (Hg^p). Of these forms, Hg^p can be effectively removed by particulate matter (PM) control devices such as electrostatic precipitators (ESPs) and fabric filters (FFs). Hg^{2+} can be easily captured by wet flue gas desulphurization systems (WFGDs) due to its high solubility in water. By contrast, Hg^0 is the most stable of these species and its residence time is estimated to be several months to one year in the atmosphere [7,8]. Moreover, Hg^0 cannot easily be removed by existing air pollution control devices (APCDs) because of its low water solubility and high volatility [9–11]. Therefore, in order to meet the increasingly stringent mercury emission limits, it is necessary to understand and analyze the distribution of Hg speciation in the flue gas from practical combustion systems.

There are three main methods for onsite mercury measurement: the Mercury Continuous Emission Monitoring System (Hg-CEMS); a wet chemistry method based on the Ontario Hydro Method (OHM); and the sorbent trap method based on the US Environmental Protection Agency (EPA) Method 30B. The Hg-CEMS is mainly used for monitoring real-time Hg emission from the stack [12]. The OHM is considered to be the standard and reference mercury speciation measurement method for flue gas but its complexity in operation and the potential for errors in

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solution preparation and analysis are non-negligible concerns [13]. Recently, the EPA Method 30B which uses chemically treated activated carbon (AC) as the required sorbent has begun to be gradually accepted worldwide. It is seen as an effective alternative to OHM because of its convenient operation, high precision and low cost [14]. However, the high price of imported sorbent traps and AC remains a problem for its wide industrial application in China.

Recently, the majority of Chinese power plants have been equipped with advanced APCDs, such as selective catalytic reduction (SCR) or selective non-catalytic reduction (SNCR), ESP or FF, WFGD with an aim to reduce the emission of nitrogen oxides (NO_x), PM and sulphur dioxide (SO_2), respectively. However, increased application of these APCDs will not only increase the production of coal combustion by-products (e.g., gypsum and WFGD wastewater), but also affect the partitioning behavior and emission of mercury. Lu et al. [15,16] found that the concentration of Hg^{2+} in the flue gas increased and the Hg^0 concentration decreased after the ESP, and chlorine in coal and unburned carbon in fly ash appear to be the primary components responsible for enhancement of mercury oxidation and capture in practical systems. Álvarez-Ayuso et al. [17,18] determined the abatement capacity of WFGD for Hg and found that the removal rate ranged from 30.4% to 78.4% and most of the Hg removed by WFGD was found in the WFGD gypsum. However, there is concern that disposal (e.g., landfill and deposition outdoors) and utilization of fly ash and gypsum (e.g., production of concrete) may have harmful impacts on the environment and human health. Thus, the understanding of partitioning and redistribution behavior of Hg across APCDs will help to develop more promising Hg emission control technologies for use in power plants.

Petroleum coke is a solid byproduct derived from the petroleum refining process [19]. As an alternative energy source to traditional fossil fuel, the production of petroleum coke has been increasing with the rapid development of heavy oil processing. However, petroleum coke was classified as an inferior and high polluting fuel because of its high ignition temperature, long burning time and high sulphur content [20,21]. Circulating fluidized bed (CFB) technology has high combustion efficiency, low pollutant emissions and excellent fuel flexibility and, thus, it appears to be an effective way to burn petroleum coke [22]. Previous studies have mainly focused on mercury migration and emission in CFB boilers when burning fuels like coal, sewage, biomass, coal gangue, etc., and there is little research on mercury emission from burning mixed petroleum coke and coal as fuel [23,24]. In order to ensure the clean and effective use of petroleum coke, it is essential to study the migration and emission of mercury in CFB boilers co-firing petroleum coke and coal.

In this paper, field tests on mercury migration and emission characteristics were carried out at three 410 t/h CFB boilers burning mixed petroleum coke and coal as fuel. The Hg concentration and speciation distribution were simultaneously sampled at both inlet and outlet of the ESP, FF and WFGD. The main objectives of this study are as follows: (1) determination of mercury mass balance and its distribution; (2) determination of mercury concentration and speciation across APCDs based on both OHM and EPA Method 30B; (3) determination of mercury removal rate from APCDs; (4) the measurement of mercury contamination to the environment and determining the emission factor.

2. Material and methods

2.1. Description of utility boilers

The migration and emission of Hg were studies at three CFB utility boilers. The detailed configurations of these boilers are shown in Table 1. The rated capacities of each tested boiler is 410 t/h. Boiler #1 comprises SNCR + ESP + WFGD while boiler #2 and #3 have SNCR + FF + WFGD in series as APCDs to control the emission of NO_x, PM and SO_2. The SNCRs use urea as the denitrification reagent. The WFGDs are typical vertical spray towers based on limestone-gypsum, which consists of a circulating pump, spray nozzles, spray layer, oxidation zone and demister. In addition to the WFGDs, limestone powder was used as an in-furnace desulphurization agent during the combustion process. Powdered limestone was added to the furnace from a limestone bin by means of pneumatic conveying. The additional use of limestone powder is necessary in order to meet the stringent SO_2 emission limit in China and it further has the potential to provide the co-benefit effect of mercury capture. The Ca/S mole ratios for the three boilers are given in Table 1.

Table 1
Configuration of tested boilers.

Item	Boiler type	Capacity/($t \cdot h^{-1}$)	APCDs	[Ca/S][b]
#1	CFB	410	SNCR + ESP + (IFD[a] + WFGD)	2.4
#2	CFB	410	SNCR + FF + (IFD[a] + WFGD)	3.2
#3	CFB	410	SNCR + FF + (IFD[a] + WFGD)	3.2

[a] IFD: in-furnace desulphurization.
[b] Ca/S mole ratio used in the fluidized bed (added limestone to fuel sulphur).

Of these boilers, boiler #1 burns 100% coal, while both boiler #2 and boiler #3 burn mixed fuel of petroleum coke and coal with a blending ratio of 1:2. The proximate and elemental analyses of the fuels are shown in Table 2. According to the National Coal Classification Standard of China (GB/T 7562-2010), the mixed fuel sample can be classified as a bituminous coal when co-firing petroleum coke. Here, the fixed carbon and sulphur contents in the mixed fuel sample (boiler #2, boiler #3) are higher than those in the coal sample (boiler #1), while the ash content in the mixed fuel sample is lower than that in the coal sample. The mercury contents in coal and mixed fuel are 0.084 mg/kg and 0.066 mg/kg, respectively. Chlorine in coal is considered to be an important factor influencing mercury partitioning behavior. In this study, the chlorine content in coal and mixed fuel is 110 mg/kg and 77 mg/kg, respectively, which is significantly lower than most of the coals used commercially in China (260 mg/kg) and the United States of American (614 mg/kg) [25].

2.2. Sampling process

During the field test, the total mercury concentration in the flue gas was sampled based on the Ontario Hydro Method (OHM) and EPA Method 30B, respectively. The mercury speciation in the flue gas was analyzed based on the OHM. Both sampling methods were tested simultaneously at three points, namely both the inlet and outlet of ESP, FF and WFGD. Detailed sampling locations and configurations for the power plant are shown in Fig. 1. The temperatures at the furnace and sampling locations are shown in Table 3.

The sampling equipment for the two sampling methods was by means of an Apex mercury instrument made in America. The flue gas sample was first extracted from the gas duct isokinetically by a probe with a quartz fiber filter maintained at 120 °C to prevent the condensation of water vapor and the adsorption of Hg vapor on the inner face of filter. The Hg^P was collected on a quartz fiber filter. In the case of the OHM, the flue gas sample subsequently flows through a series of impingers placed in an ice bath. The Hg^{2+} was collected by the first three impingers containing 1 mol/dm^3 KCl solution, and Hg^0 was collected in the fourth impinger containing 5% V/V H_2O_2-10% V/V HNO_3 solution and three impingers with a solution of 4% W/V $KMnO_4$-10% V/V H_2SO_4. The eighth impinge, containing silica gel, was used to remove the moisture from the previous impinger train before entering the following auxiliary equipment such as thermometer, vacuum gauge, air-tight pump, gas metering console, etc.

For EPA Method 30B, the flue gas sample after filtration subsequently flowed through paired traps filled with potassium iodide-treated activated carbon (AC-KI) to capture the gaseous mercury. The AC-KI sorbent trap was procured from US Ohio Lumex Inc., which is

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