



Research article

Partitioning behavior of Arsenic in circulating fluidized bed boilers co-firing petroleum coke and coal

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ABSTRACT

The emission of Arsenic from coal-fired power plants has generated widespread environmental and human health concerns. This paper discusses Arsenic partitioning from three 440 t/h circulating fluidized bed (CFB) boilers co-firing petroleum coke and coal. All the boilers were equipped with electrostatic precipitator (ESP) or fabric filter (FF), and wet flue gas desulfurization (WFGD). Flue gas was sampled simultaneously both up- and down-stream of the ESP/FF and at the outlet of the WFGD based on EPA Method 29. Concurrent with flue gas sampling, feed fuel, bottom ash, ESP/FF ash, WFGD gypsum, WFGD wastewater, limestone slurry and flush water were also collected. The results show that, for three tested CFB boilers, the overall mass balance ratios of As ranged from 80.0%–114.2%, which can be considered to be acceptable and reliable. Most of the As was distributed in the bottom ash and ESP/FF ash with the values of 17.4%–37.5% and 55.6%–77.5%, respectively. Speciation analysis suggests that As^{5+} was the major water-soluble species in the feed fuel, bottom ash and fly ash, while As^{3+} was found to be the dominant species in WFGD wastewater. For three CFB boilers, the concentrations of total As in the stack emission were 0.97, 0.32 and 0.31 $\mu\text{g}/\text{m}^3$, respectively. The CFB boiler equipped with ESP/FF + WFGD was shown to be able to provide good control of the emission of As emitted into the atmosphere.

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1. Introduction

Arsenic (As) is a hazardous metalloid elements, and can be released to the environment during coal combustion, municipal solid waste incineration and metal smelting [1]. Coal combustion is the major source of power generation in China. It has been reported that in China, nearly 50% of coal consumption was for electric power and heat generation [2]. Despite low As concentration in the majority of coals, the total amount of As released during coal combustion is considerable because of the substantial amounts of coal consumed annually [3,4]. Presently, coal-fired power plants are recognized as the major anthropogenic As emission source [5,6].

When As is introduced into the environment during the coal combustion process, it poses serious pollution problems for environment, as well as a danger to human health. It is reported that for people who live in endemic areas with high As concentrations in drinking water or in regions where coal burning is important, long term exposure to As causes acute and chronic adverse effects including cancer [7,8]. The toxicity of As is highly dependent on its chemical form [9,10]. In general, inorganic arsenite (As^{3+}) is a more potent toxicity than the inorganic arsenate (As^{5+}) [7,11]. Numerous studies have investigated the As

speciation including (As^{3+} and As^{5+}) in fly ash [9,12,13]. However, information on the speciation of As in combustion byproducts such as bottom ash, gypsum and wastewater from power plants is scarce.

In the combustion zone, As and its compounds that are associated with coal's mineral matter and organic matter will mostly volatilize, but some of it will be retained in bottom ash. As the flue gas cools down, a portion of gaseous As and its compounds will condense on the surface of particles and recombine with particulates through physical absorption and chemical reaction [14–16]. The As fraction absorbed on the fine particles may be removed by conventional air pollution control devices (APCDs) such as electrostatic precipitators (ESP) or fabric filters (FF), and wet flue gas desulfurization (WFGD), etc. In general, As will be emitted into the atmosphere in both in gaseous form and particulate bound. In recent years, extensive field tests on the partitioning behavior of As across APCDs in coal-fired power plants have been conducted by many researchers [14,17,18]. Cheng et al. [19] carried out a field test on a coal fired power plant to investigation the distribution of As for selective catalytic reduction (SCR), ESP, and WFGD. Swanson et al. [20] determine the partitioning of As in combustion by products including bottom ash, economizer ash and fly ash from two US power plants. However, there is a dearth of studies on As partitioning behavior across APCDs in circulating fluidized bed (CFB) boilers.

Petroleum coke is a byproduct from the oil refining process, which possesses high heating value, high fixed carbon content, high sulfur

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content, and low volatiles content [21]. Due to its excellent fuel flexibility and combustion efficiency, the use of CFB combustion technology for co-firing petroleum coke and coal has been steadily increasing [22,23]. Currently, studies concerning As emission characteristics in CFB boilers have focused mainly on burning or gasifying fuels like biomass and municipal solid waste, and there is little published research on As emission from petroleum coke and coal co-firing [24–26]. Given that petroleum coke is considered to be a significant alternative energy source, it is essential to study As partitioning behavior and environmental effects in CFB boilers co-firing petroleum coke and coal.

The application of APCDs in the power plant can increase the amount of coal combustion byproduct (e.g., fly ash, gypsum) and affect the redistribution behavior of As in some of these combustion byproducts. Furthermore, the disposal (e.g., landfilling and storage) and utilization (e.g., construction) of fly ash and WFGD gypsum, as well as discharge of WFGD wastewater, potentially pose a threat to humans, both in the soil and groundwater [27,28]. To study the partitioning behavior and environmental impacts of As, onsite tests were conducted at three CFB boilers equipped with ESP/FF and WFGD. Flue gas sampling for As was carried out at three points simultaneously, based on the U.S. Environmental Protection Agency (EPA) method 29. The main objectives of this study were to determine: (1) the concentration and enrichment of As in feed fuel and combustion byproducts; (2) mass balance ratio and distribution of As in the entire system; (3) removal efficiency of As by APCDs; and (4) environmental impact of As.

2. Materials and methods

2.1. Boiler description

The study was conducted at three CFB boilers (unit #1, unit #2, unit #3) co-firing petroleum coke and coal in Guangdong province, China. Each boiler had a rated capacity of 440 t/h. The detailed configurations of these boilers are provided in Table 1. Selective non-catalytic reduction (SNCR) units were employed to reduce nitrogen oxide emissions and the ESP and FF were used for particulate matter (PM) removal. The WFGD unit was operated using limestone as reagent. In addition to WFGD, limestone powder was used as furnace desulphurization agent during the combustion process. Powdered limestone was added to the furnace from limestone bin by means of pneumatic conveying. The additional use of limestone powder is necessary in order to meet stringent SO₂ emission limit and it further has a potential to provide the co-benefit effect of As capture. The Ca/S mole ratios for three units are given in Table 1. Unit #1 was fueled with 100% bituminous coal, while both unit #2 and unit #3 were fueled with the same coal used in unit #1 and petroleum coke with a blending ratio of 2:1. Boiler conditions were maintained as stable as possible during the sampling process. Proximate and ultimate analyses of the fuels are given in Table 2, are based on the National Coal Classification Standard of China (GB/T 5751-2009). The analysis methods used for each test are given in Table 3.

2.2. Sampling procedure

For the flue gas sampling, a sampling run was simultaneously begun at all sampling locations. Detailed simultaneous sampling locations in

the tested boilers are presented in Fig. 1. The emission of As from flue gas was determined by EPA method 29 [29]. This method has been historically the most commonly used method for determining mercury and other trace metal elements including As. Gas samples were collected from three sampling points along the flue gas path, at the inlets/outlets of ESP/FF, WFGD. For a given sampling location, gas samples were withdrawn isokinetically from the flue gas through a probe with a quartz fiber filter, maintaining the temperature at 120 ± 20 °C. The gaseous form of As was absorbed and retained by 5% (v/v) nitric acid (HNO₃)/10% hydrogen peroxide (H₂O₂) in two ice-bath-chilled impingers. The particulate bound As can be captured by means of a quartz fiber filter. Samples in both the quartz fiber filter and the HNO₃/H₂O₂ solutions were collected and recovered immediately after collection. The whole sampling process lasted for 2 h using a U.S. Apex mercury instrument (Apex Instruments, Fuquay-Varina, NC, USA).

For solid and liquid materials, feed fuel, bottom ash, ESP/FF ash and WFGD samples were collected simultaneously in step with flue gas sampling every hour. The configuration of the WFGD system and locations of the WFGD samples are shown in Fig. 2. The WFGD samples which consisted of input streams such as limestone slurry and flush water, and output streams such as WFGD gypsum and WFGD wastewater were collected from the appropriate locations from WFGD system. The WFGD gypsum samples were collected during the test period when the dewatering system was in operation. Solid samples were stored in zip-lock bags and liquid samples were preserved in high boron silicon bottles and transported to the chemical laboratory immediately after sampling for immediate As analysis. In addition, the temperature at furnace and flue gas locations are given in Table 4. All sampling tests were carried out twice to obtain duplicate results and reduce experimental uncertainties. The results presented in this paper are average values.

2.3. Elemental analysis

The solid samples including feed fuel, bottom ash, ESP/FF ash, particulate matter in the flue gas and WFGD gypsum were firstly dried in the oven at 50 °C for 8 h, and then milled and sieved to 200 meshes. The pulverized samples were digested using an acid mixture (HNO₃:HCl:HF = 3:1:1) in a microwave oven. Then the As concentration in the digestion solutions was determined by inductively-coupled plasma mass spectrometry (ICP-MS). For WFGD limestone slurry and WFGD wastewater, which are not clear or transparent, the solid and liquid portions were separated through filtration and drying, and then the total As content in them was obtained by calculating the content in solid and liquid portion. As content in WFGD flush water, which is clear, can be determined by ICP-MS directly. The detection limits of As in solid and liquid samples by ICP-MS are 0.1 mg/kg and 0.05 µg/L, respectively.

All measurements of As concentration were carried out in third times to obtain parallel results and the relative standard deviation of parallel results is <3%.

2.4. Speciation analysis

Water-soluble As³⁺ and As⁵⁺ content in bottom ash, ESP/FF ash, WFGD wastewater and WFGD gypsum were determined using ultrasonic microwave-assisted extraction followed by high-performance liquid chromatography coupled with ICP-MS (HPLC-ICP-MS). This method has been used successfully in several previous studies [30,31]. Here, 10-mg solid samples were weighed into HDPE bottles and then 10 mL 1% (v/v) HNO₃ was added. After ultrasonic microwave-assisted extraction for 40 min with constant agitation during the extraction process, samples were centrifuged at 7000 rpm for 10 min and then filtered through 0.25 µm PVDF filter. For liquid samples, As³⁺ and As⁵⁺ content in clear samples can be directly determined by HPLC-ICP-MS after filtration. The accuracy of As³⁺ and As⁵⁺ content in samples was determined using a blending solution of GBW08667 and GBW08666 as standard reference

Table 1
Configuration of tested boilers.

Item	Rated capacity (t/h)	APCDs	Fuel type	[Ca/S] ^a
Unit #1	440	SNCR + ESP + WFGD	100% coal	2.8
Unit #2	440	SNCR + FF + WFGD	66.7% coal + 33.3% PC	4.8
Unit #3	440	SNCR + FF + WFGD	66.7% coal + 33.3% PC	4.8

^a Ca/S mole ratio used in the fluidized bed (added limestone to fuel sulfur).

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