



Emission characteristics for gaseous- and size-segregated particulate PAHs in coal combustion flue gas from circulating fluidized bed (CFB) boiler[☆]

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ABSTRACT

The partitioning behavior of polycyclic aromatic hydrocarbons (PAHs) between gaseous and particulate phases from coal-fired power plants (CFPPs) is critically important to predict PAH removal by dust control devices. In this study, 16 US-EPA priority PAHs in gaseous and size-segregated particulate phases at the inlet and outlet of the fabric filter unit (FFs) of a circulating fluidized bed (CFB) boiler were analyzed. The partitioning mechanisms of PAHs between gaseous and particulate phases and in particles of different size classes were investigated. We found that the removal efficiencies of PAHs are 45.59% and 70.67–89.06% for gaseous and particulate phases, respectively. The gaseous phase mainly contains low molecular weight (LMW) PAHs (2- and 3-ring PAHs), which is quite different from the particulate phase that mainly contains medium and high molecular weight (MMW and HMW) PAHs (4- to 6-ring PAHs). The fractions of LMW PAHs show a declining trend with the decrease of particle size. The gas-particle partitioning of PAHs is primarily controlled by organic carbon absorption, in addition, it has a clear dependence on the particle sizes. Plot of $\log(\text{TPAH}/\text{PM})$ against $\log D_p$ shows that all slope values were below -1 , suggesting that PAHs were mainly adsorbed to particulates. The adsorption effect of PAHs in size-segregated PMs for HMW PAHs is more evident than LMW PAHs. The particle size distributions (PSDs) of individual PAHs show that most of PAHs exhibit bi-model structures, with one mode peaking in the accumulation size range (2.1–1.1 μm) and another mode peaking in coarse size range (5.8–4.7 μm). The intensities of these two peaks vary in function of ring number of PAHs, which is likely attributed to Kelvin effect that the less volatile HMW PAH species preferentially condense onto the finer particulates. The emission factor of PAHs was calculated as 3.53 mg/kg of coal burned, with overall mean EF_{PAH} of 0.55 and 2.98 mg/kg for gaseous and particulate phase, respectively. Moreover, the average emission amount of PAHs for the investigated CFPP was 1016.6 g/day and 371073.6 g/y, respectively.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous global contaminants and are potentially carcinogenic and mutagenic to human beings (Meyer et al., 2013; Wang et al., 2017). The majority

of PAHs in modern environmental compartments come from incomplete combustion of carbonaceous materials during power generation and industrial production processes (Yunker et al., 2011; Kim et al., 2013; Wang et al., 2015a; b). Global total emissions of 16 U.S. Environmental Protection Agency (US-EPA) priority PAHs was 504 Gg/year in 2007, with over 50% of these emissions coming from South, East and Southeast Asia (Shen et al., 2013a). Coal combustion in coal-fired power plants (CFPPs) has become the major contributor of PAHs and particulate matters (PMs) in the atmosphere of China (Song et al., 2015; Wang et al., 2016; Guo et al., 2017), where

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approximately 47% of coal production and consumption (3.87 and 4.12 billion tons in 2014, respectively) is used for electricity generation during 2010–2014 (NBSC, 2016).

The main boiler types in Chinese CFPPs include pulverized coal-fired (PC) boiler, coal stoker and circulating fluidized bed (CFB) boiler, PC boilers are the most common, accounting for over 85% of coal-fired boilers in China (Tian et al., 2014). Although the share of CFB boilers is small, they play a vital role in generating power from low quality coal, lignite and alternative fuels (e.g., biomass, municipal wastes). In addition, CFB boilers are characterized by low NO_x emissions due to low combustion temperature, and low SO₂ emissions because of in situ desulfurization (Gómez et al., 2014). Up to now, there are ~6000 CFB boilers are under operation in China, with electricity output of over 70 GWe, or of ~10% of the total electricity generation from CFPPs (NBSC, 2016). During the past decades, the status of Chinese CFPPs has changed substantially to confront the pollutant emissions from CFPPs, including the replacement of small units with low combustion efficiency by ones larger than 300 MW and the increasing installation of conventional air pollution control devices (APCDs), such as selective catalytic reduction denitration device (SCR), electrostatic precipitators (ESPs) and wet flue gas desulfurization (WFGD), etc. In addition, Chinese government has enacted strict regulations (e.g., Emission Standard of Air Pollutants for Thermal Power Plants (ESAPTPP, GB 13223–2011) in recent years to meet the ultralow-emission requirement for pollutant emission. As a result of the wide application of advanced APCDs and management strategies, the national total emissions of PM, SO₂, NO_x and hazardous trace elements have shown a gradual decline since the last ten years (Zhao et al., 2008; Tian et al., 2013, 2014).

Field tests have demonstrated that the application of conventional APCDs has significant impacts on the cooperative reduction of PAHs existing in flue gases. Lee et al. (2002) indicated that the removal efficiency of high molecular weight (HMW) PAHs by the ESP used in medical waste incinerators (MWIs) with a mechanical grate is only 2.2%, whereas the removal efficiency is 28.8% for HMW PAHs by the ESP used in MWIs with a fixed grate. Yin et al. (2007) and Guerriero et al. (2008) indicated that the removal efficiency of PAHs by de-dust devices are very variable, ranging from 5.2% for ESP to 59.5% for wet scrubber. WFGD has been shown to have a significant co-benefit of PAH abatement in both particulate and gaseous phases, especially for the HMW PAHs (Wang et al., 2013, 2015a, 2016). The total concentration of 16 US-EPA PAHs reduced from 6.34 µg/m³ (at the inlet of WFGD) to 0.87 µg/m³ (at the outlet of WFGD) during the flue gas desulfurization. Hsu et al. (2016) indicated the overall PAH removal efficiency that is achieved with SCR + ESP + FGD is 74.7%, and HMW PAHs have a lower removal efficiency than the other PAHs by ESP due to the condensation of PAHs on the particulate surface across the ESP as the cooling-down of flue gas temperature. A recent study reported by Li et al. (2016) indicated that the removal efficiencies of the low-temperature ESP (LLT-ESP) on PM_{2.5} and PM associated PAHs are 86% and 87%, respectively.

However, it has been indicated that APCDs are inefficient for capturing gaseous PAHs and PAHs bound to fine/ultrafine particles (Hsu et al., 2016; Mastral and Callén, 2000; Xu et al., 2016a, 2016b; Zhang et al., 2016). Therefore, large amounts of PAHs have been emitted into the atmosphere along with coal combustion flue gas (Kim et al., 2013; Mu et al., 2014). Knowledge on the PAH partitioning behaviors between gaseous and particulate phases and in size-segregated PMs, and their variations across APCDs is critically important to predict PAH emissions. Previous studies have shown that PAH partitioning between gaseous and particulate phases is primarily dependent on their volatilities, and that PAH partitioning in the particulates is largely dependent on the size, unburned

carbon content and metal species of particulates (Yang et al., 1998, 2002; Arditoglou et al., 2004; Zhou et al., 2009; Shen et al., 2011; Pergal et al., 2013; Kong et al., 2011, 2013; Hu et al., 2014). However, the partitioning behavior of PAHs has not been well constrained, especially for the coal combustion in CFB boilers. In this study, the variation characteristics of PAHs in both gaseous and particulate phases are investigated in a CFB boiler where the flue gases were simultaneously sampled before and after the fabric filters units (FFs). The objectives of the current study is to (1) investigate the distributions of PAHs in size-segregated PMs and gaseous phases, (2) study the partitioning mechanisms of PAHs between gaseous and particulate phases and in size-segregated PMs; (3) establish the emission factors of PAHs.

2. Materials and methods

2.1. Configuration and operation of the studied CFB boiler

The field measurement was carried out at a 15 MW circulating fluidized bed (CFB) boiler in Hefei city, China. The sketch of this unit and the sampling locations are illustrated in Supporting Information Fig. S1. The CFB boiler was equipped with a limestone injection system for SO₂ control and a urea injection system for NO_x control and fabric filters units (FFs) for removing PMs from flue gases. Crushed bituminous coals with particle size less than 10 mm were fed into and burned in the furnace. The feed coals was from the Permo-Carboniferous coal deposits of Huainan coalfield, which are characterized by low-medium volatile matters, medium sulfur content, medium calorific value and ash yield. The generated combustion flue gas first enter into the cyclones installed downstream of the furnace, where the PMs of large size are separated out and recycled into the furnace through a U-shaped refeeder. The remaining flue gas comprising of fine PMs passed through the economizer and air preheater successively before entering into the fabric filter units (FFs). The operating temperature in combustion zone of the boiler was ca. 850 °C, and decreased to 136.3 ± 3.7 °C before the FFs and to 117.5 ± 3.2 °C after the FFs. The flue gas flow velocity were measured as 9.6 ± 1.7 m/s and 10.7 ± 1.4 m/s at the inlet and outlet of FFs, respectively.

2.2. Sample collection

The sampling apparatus for gaseous and particulate PAHs was modified after the U.S. EPA Method 23A (Fig. 1). The flue gas was pumped through a sampling probe, and then passed over a modified size-segregated impactor for collecting PMs on different quartz fiber filters (Whatman International, Maidstone, UK, φ 81 mm) and an XAD-2 polystyrene resin (SUPELCO, Amberlite) for collecting gaseous PAHs. Herein a modified eight-stage non-viable impactor

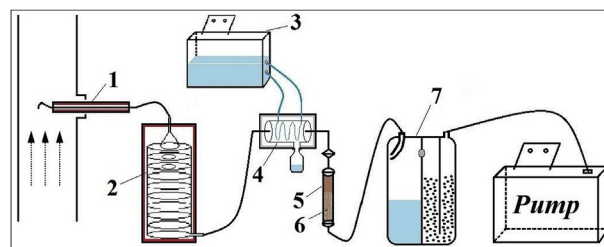


Fig. 1. Schematic diagram sampling apparatus for collecting size-segregated PMs and gaseous PAHs. 1) sampling probe; 2) modified Anderson eight-stage non-viable impactor; 3) cooling-water machine; 4) glass condenser; 5) brown glass trap; 6) XAD-2 resin; 7) gas-water separator. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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