



Characterization of PAHs within PM₁₀ fraction for ashes from coke production, iron smelt, heating station and power plant stacks in Liaoning Province, China

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ABSTRACT

Polycyclic aromatic hydrocarbons within PM₁₀ fraction of ashes from two coke production plants, one iron smelt plant, one heating station and one power plant were analyzed with GC–MS technique in 2009. The sum of 17 selected PAHs varied from 290.20 to 7055.72 µg/g and the amounts of carcinogenic PAHs were between 140.33 and 3345.46 µg/g. The most toxic ash was from the coke production plants and then from the iron smelt plant, coal-fired power plant and heating station according to BaP-based toxic equivalent factor (BaP_{eq}) and BaP-based equivalent carcinogenic power (BaP_{eq}). PAHs profile of the iron smelt ash was significantly different from others with coefficient of divergence value higher than 0.40. Indicatory PAHs for coke production plants, heating station and coal-fired power plant were mainly 3-ring species such as Acy, Fl and Ace. While for iron smelt plant, they were Chr and BbF. Diagnostic ratios including Ant/(Ant + Phe), Flu/(Flu + Pyr), BaA/Chr, BbF/BkF, Ind/BghiP, IND/(IND + BghiP), BaP/BghiP, BaP/COR, Pyr/BaP, BaA/(BaA + Chr), BaA/BaP and BaP/(BaP + Chr) were calculated which were mostly different from other stacks for the iron smelt plant.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds that consist of two or more fused aromatic rings with some of them being carcinogenic and mutagenic (Yang et al., 2002; Sun et al., 2006). PAHs emitted from anthropogenic sources can be divided into stationary combustion sources and mobile combustion sources like industrial production, waste incineration, transportation and so on (Yang et al., 1998; Mastral and Callén, 2000; Yang et al., 2002). They are generated by incomplete combustion and pyrolysis or pyro-synthesis of fossil fuels and other organic matters which represent a risk to both environment and health (Mastral et al., 1996; Yang et al., 1998; Liu et al., 2000; Mastral and Callén, 2000; Mastral et al., 2001; Morawska and Zhang, 2002; Johansson and Bavel, 2003a; Santos et al., 2004; Sun et al., 2005; Chao et al., 2008; Ravindra et al., 2008). During coal combustion, PAHs were found in both flue gas and fly ash (Sun et al., 2006; Chao et al., 2008). There were several publications concerning PAHs characteristic in municipal solid waste incineration bottom ash

(Johansson and Bavel, 2003a; Chou et al., 2008), industrial stack gas (Yang et al., 2002; Tsai et al., 2007; Ravindra et al., 2008; You, 2008), lime spray dryer (LSD) ash (Sun et al., 2005, 2006) and animal carcass waste incinerator ash (Chen et al., 2003) whereas data related to industrial boilers such as power plant (Arditsoglou et al., 2004), iron smelt, coke production and heating purpose boilers (Johansson and Bavel, 2003a) were still limited owing to the difficulty of the stack sampling work (Li et al., 1999). Meanwhile the quantities and characteristics of PAHs emitted from industrial boilers may vary over a wide range due to the type of input (fuel, additive etc.), the manufacturing process, the air pollution control devices, etc. (Mastral et al., 1996; Yang et al., 1998; Li et al., 1999; Manoli et al., 2004; Ahmaruzzaman, 2010). It was well recognized that fine particles of ashes has larger surface area per unit mass than coarse particles, thus more condensed PAHs can be absorbed (Liu et al., 2000; Mastral and Callén, 2000; You, 2008). However, former studies focused mainly on particles with diameters larger than 100 µm (Liu et al., 2000, 2002; Kuo et al., 2003; Arditsoglou et al., 2004; Sun et al., 2005). To our knowledge, few studies have given the PAHs concentrations in PM₁₀ fractions (particles with diameters less than 10 µm) of ashes which may be more easily inhaled by human. Therefore, it is necessary to investigate the PAHs concentrations in this fraction to fill this gap.

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From the view of utilization, ashes can be used in construction of roads, cement manufacture, building materials and low-cost adsorbent for gas and water treatment (Johansson and Bavel, 2003a; Ahmaruzzaman, 2010). The knowledge of organic environmental pollutants in ashes was scarce which limited the large-scale utilization of them (Johansson and Bavel, 2003a). Therefore, an investigation of PAHs (e.g., concentration, fate) in ashes was necessary to determine environmentally safe use or disposal options (Sun et al., 2006). PAHs adsorbed in fly ash can evaporate into the environment (Liu et al., 2000, 2002; Arditoglou et al., 2004) or enter into atmosphere by re-suspension and then diffuse far away to cause atmospheric pollution (Arditoglou et al., 2004). Coal consumption was approximately 64 Mt leading to fly ash production of 0.27 Mt in the year of 2008 in Liaoning Province and the ashes were always piled up with poor covering. So considerable amounts of PAHs within fine fraction of ashes may re-suspend into the air and cause local or regional pollution of atmospheric particulate matter.

Furthermore, to understand the relationship between sources and observed air quality, chemical mass balance (CMB) receptor model has been widely used which needs representative and reliable source fingerprints as one of its input data (Khalili et al., 1995; Tsai et al., 2007). However, source profiles for PAHs in fly ashes of PM₁₀ fraction were still limited (Yang et al., 2002; Manoli et al., 2004). Meanwhile the profiles for a specific region need update timely according to the variation of sources. The most cost effective and practical method for establishing chemical profiles of fly ash was grabbing ash samples from storage pipes, sieving them and then collecting them onto filters with a re-suspension chamber, although large industrial stack emissions require diluted sampling (Watson et al., 2002).

Considering the fact listed above, ashes from four kinds of industrial stacks including two coke production plants, one iron smelt plant, one heating station and one power plant from Liaoning Province, China were studied regarding their composition of PAHs. The purpose of this study was to: (1) determine PAHs levels in ashes from the four types of industrial plants; (2) compare the potential toxicity of PAHs emitted from these industrial stacks; (3) establish PM₁₀-bound PAHs profiles, analyze source markers and study the profiles diversities; (4) calculate diagnostic ratios of characteristic PAHs species for source identification purpose.

2. Methodology

2.1. Sampling description

Eight ash samples were collected from July to September in 2009 in Liaoning Province which was the traditionary heavy industrial base of China. Samples were collected from the storage

piles or bottom of electrostatic precipitators (Watson et al., 2002) with a plastic brush. Then ashes were wrapped using aluminum foil before sealed in labeled envelopes. About 1 kg ashes were collected for each type. Sampling sites and descriptions were shown in Table 1. For each type of dust, three samples were obtained at different locations.

2.2. Sample preparation

Ash samples were pre-treated using re-suspension method to collect particles with diameters less than 10 µm (Chow et al., 1994; Han et al., 2009). Each sample was weighed after being dried in a vacuum freeze dryer to remove the moisture. After drying, the samples were sieved through a 160-mesh screen to remove fibers and other larger particles. About 0.5 g sieved samples were suspended in a re-suspension chamber (Fig. 1) and sampled through PM₁₀ inlets with a flow rate of 20 L/min for about 2 min onto quartz-fiber filters. Filters were weighed both before and after sampling by a sensitive microbalance (Mettler M5). The balance sensitivity was ±0.001 mg.

2.3. PAHs determination

For PAHs analysis, the filters were extracted ultrasonically with dichloromethane, then concentrated using a rotary evaporator, purified with a silica gel cleanup technique and re-concentrated by rotary evaporation. Finally they were condensed to exactly 1 mL under a gentle nitrogen stream in 60 °C water bath. The extracts were transferred into two ampoule bottles and stored in a refrigerator until analysis. Gas chromatography coupled to mass spectrometry (GC–MS) was used according to the EPA Method TO-13A. For each sample, the procedures of sampling, pretreatment and analysis had been completed within one month.

PAHs in the final extracts were analyzed with a trace 2000 GC–MS (Thermo Finnigan, USA) apparatus with selected ion monitoring (SIM). Capillary column DB5–MS (length: 30 m; inner diameter: 0.25 mm; thickness: 0.25 µm) was used. Temperature increasing program at working conditions of the GC–MS was as follows: 70 °C isotherm for 2 min, 10 °C/min till 260 °C and isotherm for 8 min, then increased at a rate of 5 °C/min to 300 °C (held for 5 min). The injector was kept at constant temperature of 280 °C. Helium with a purity of 99.999% was used as carrier gas at a constant flow of 1.0 ml/min. The transfer line was heated at 280 °C. A 1 µL volume was injected by applying a hot splitless injection technique at 280 °C. The mass spectrometry was operated in electron ionisation (EI) mode with an ion source temperature of 250 °C and the electron impact energy was set at 70 eV. Response factors of the PAHs standard solution were used to calculate the masses of sampled

Table 1
Sampling sites and descriptions.

Locations	Fuel types	S (%)	Ash (%)	H ₂ O (%)	V (%)	calorific value (kJ/kg)	No.	Plant types	collection location for ash samples	Mnemonic
Benxi integrated iron and steel plant, Benxi city	Raw coal	0.9	25	0.7	46	17 896	1	Coke making	coking processes storage piles	BCCPS
							2	Coke making	bottom of electrostatic precipitator	BCBEP
							3	iron smelt	bottom of electrostatic precipitator	BIBEP
							4	heating	bottom of electrostatic precipitator	SHBEP
Shendong heating station, Shenyang city	Raw coal	0.56	27	0.9	45	18 634				
Nenggang power plant, Fushun city,	Raw coal	0.5	28.8	0.9	47.5	15 920	5	heating	ash storage house after electrostatic precipitator and wet FGD	NPASH
Anshan integrated iron and steel plant, Anshan city,	62% coking coal, 15% fat coal, 23% meagre coal	0.69	10	10	24–31	/	6	Coke making	coking processes storage piles	ACCPs
							7	Coke making	the stack before electrostatic precipitator	ACBEP
							8	Coke making	the bottom of electrostatic precipitator	ACSEP

V: volatile portion.

/: not detected.

FGD: flue gas desulphurization.

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