



Seasonal variations and source apportionment of complex polycyclic aromatic hydrocarbon mixtures in particulate matter in an electronic waste and urban area in South China



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HIGHLIGHTS

- Air MW < 302 PAH concentrations were not significantly different at two sites.
- E-waste recycling is a more significant source of MW 302 and halogenated PAHs.
- Coal combustion, biomass burning, and vehicular emission were the major sources.

GRAPHICAL ABSTRACT



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ABSTRACT

Complex polycyclic aromatic hydrocarbon (PAH) mixtures including parent PAHs, high molecular weight PAHs (MW 302 PAHs), and halogenated PAHs (HPAHs) were measured in particulate matter (PM) in an urban area and a rural electronic waste area in South China. The concentrations of MW < 302 PAHs at two sites were not significantly different with annual means of 23.2 ± 17.2 and 33.7 ± 29.0 ng/m³, respectively. However the concentrations of both MW 302 PAHs (5.35 ± 3.72 ng/m³) and HPAH (49.9 pg/m³) were significantly higher at the e-waste site than the urban site (2.81 ± 2.36 ng/m³ and 28.2 ± 28.5 pg/m³), suggesting e-waste recycling being a significant source of these PAHs. The majority of PAHs exhibited higher concentrations in winter and spring and lower concentrations in fall and summer. Meteorological conditions and increased emissions of PAHs in northern China due to domestic heating in colder seasons are important factors influencing the PAH seasonal variations. Source apportionment by the chemical mass balance (CMB) model indicated that residential stoves (coal combustion), industrial boilers (coal combustion), biomass burning, and vehicular emission accounted for $38 \pm 14\%$, $30 \pm 11\%$, $22 \pm 22\%$, and $10 \pm 7\%$ of the PAHs in the urban PM, respectively. Comparable contributions from

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these sources were also observed for PM at the e-waste site. PAH emission factors are needed for primitive e-waste recycling to further understand the importance of this source to ambient air.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic contaminants that are formed mainly from incomplete combustion or pyrolysis of organic matter such as fossil fuel, wood, and municipal waste (Shen et al., 2010). Although they have been extensively studied, PAHs in the environment still attract great attention because PAHs are ubiquitous in both urban and rural environments and many of them are mutagenic and carcinogens (Armstrong et al., 2004). Perera et al. found that prenatal exposure to airborne PAHs can adversely affect child behavior (Perera et al., 2012). A study from Chongqing, China indicated that environmental concentration of PAH exposure is associated with increased sperm DNA damage (Han et al., 2011). In addition, substituted PAH derivatives (e.g., nitrated, oxygenated, and halogenated PAHs) and high molecular-weight PAHs (MW 302 Da) PAHs have also attracted concern in recent years (Jariyasopit et al., 2014; Reisen and Arey, 2005; Zimmermann et al., 2013). These PAHs have been found in municipal waste incinerator gas and various environmental matrices (Tsapakis and Stephanou, 2007; Watanabe and Noma, 2009; Witter and Nguyen, 2016), and some of them are identified to be more persistent, mutagenic, and carcinogenic compared to parent PAHs (Kawanaka et al., 2008; Ohura et al., 2009).

Due to the rapid economic development and huge population, China consumes significant amount of energy for industry, transportation, and domestic combustion, and resulted in substantial emissions of PAHs to the atmosphere (Zhang et al., 2007). It was estimated that China emitted 21% of the global atmospheric emission (Shen et al., 2013). The Pearl River Delta (PRD), located in Guangdong province, South China, is one of the most highly urbanized and industrialized regions in China. The PAH emission density in the PRD region was reported to be much higher than the average PAH emission density for China (Zhang et al., 2007). The contamination of PAHs in the urban air of the PRD region has been reported in previous studies (Bi et al., 2003; Gao et al., 2012; Li et al., 2006; Liu et al., 2015; Yang et al., 2010). These studies indicated that the PAH concentrations elevated significantly in winter season and coal combustion and vehicular emission were the main sources of PAHs in the urban air (Li et al., 2006; Yang et al., 2010; Liu et al., 2015). Nevertheless, in most of these studies, the samples were collected in short periods (one or two seasons), whereas year-round data are limited. Furthermore, the PAH sources in the PRD air are largely identified using molecular diagnostic ratios but quantitative information on the sources is rare (Gao et al., 2013). A large electronic waste (e-waste) recycling site is located in the rural area adjacent to the PRD. Primitive e-waste recycling has been proved to be significant source of PAHs to the ambient environment (Ma et al., 2009; Wang et al., 2012b; Wei et al., 2012).

Particulate matter (PM) samples were collected year round from an urban site in Guangzhou and an e-waste site in a rural area in South China and analyzed for complex PAH mixtures including 32 MW < 302 PAHs, 16 MW 302 PAHs, and 27 halogenated (16 chlorinated and 11 brominated) PAHs (HPAHs). In a previous study, we reported the benzo[a]pyrene equivalent (BaP_{eq}) concentration of these PAHs in the PM and the associated human inhalation cancer risk (Wang et al., 2012a). In this study, we aim to understand the seasonal variations and spatial differences in the contamination of these PAHs and the characteristics of PM in the air at the two sites where distinct sources may exist. The chemical mass balance (CMB) model is performed to conduct source apportionment of PAHs in the air. To our knowledge, this is the

first effort to compare the simultaneous air pollution of PAHs at urban and e-waste sites.

2. Materials and methods

The detailed materials and methods have been given in a previous study (Wang et al., 2012a) and are described briefly here.

2.1. Sampling

The urban area air sampling site was located in Guangzhou, the capital of Guangdong province and the largest city in South China (Fig. S1 in the Supplementary Material). The samples were collected from the rooftop of building (approximately 20 m tall) on the Guangzhou Institute of Geochemistry campus. This site is representative of residential areas in Guangzhou without major industrial sources nearby. The e-waste area air sampling site is approximately 70 km north of the urban site, which is one of the three largest e-waste recycling sites in China. The e-waste recycling processing is concentrated mainly in a number of villages in two towns with an area of 330 km². The samples were collected from the rooftop of a building (10 m above the ground) in a small village.

Total suspended particles were collected on quartz fiber filters for 24 h using a high volume air sampler on five consecutive days of each month. The sampling was conducted simultaneously at the e-waste and urban sites from May 2010 to April 2011 and 60 samples were obtained, respectively. After sampling, the filters were wrapped in aluminum foil, sealed in polyethylene bags, returned to the lab, and stored at –20 °C until analysis.

2.2. Analysis methods

PM mass was weighed using a balance with accuracy of 0.01 mg after the filters were conditioned in a desiccator for 24 h. A small sample punch (1 × 1.5 cm²) was taken from the quartz filters to determine the contents of organic carbon (OC) and elemental carbon (EC) using National Institute of Occupational Safety and Health (NIOSH) 5040 method by an OC-EC aerosol analyzer (Sunset Laboratory Inc., US) (Birch and Cary, 1996).

The PM-loaded filters were spiked with recovery standards and extracted with a 1:1 mixture of hexane and acetone using pressurized liquid extraction (ASE300, Dionex Corp., Sunnyvale, CA, USA). The extracts were solvent exchanged to hexane and concentrated to 1 mL using Turbovap II (Biotage, Charlotte, NC, USA). The extracts were further purified with 20 g silica solid phase extraction (SPE) columns (Varian, Inc., Santa Clara, CA). The eluates were spiked with internal standards (*d*₁₀-acdnaphthene, *d*₁₀-fluoranthene, *d*₁₂-benzo[k]fluoranthene, and ¹³C₁₂-PCB138), and concentrated to 1 mL for analysis.

The target compounds were listed in Table S1. The parent PAHs and MW 302 PAHs were analyzed using an Agilent 5890 gas chromatograph (GC) (Agilent Technologies) coupled to an Agilent 5973 mass spectrometer (MS) operated in selected ion monitoring mode and using electron impact ionization. The HPAHs were analyzed using the same instrument operated in negative chemical ionization mode. Measurement of the HPAHs was achieved using a DB-5MS (30 m × 0.25 mm i.d., 0.25 μm film thickness) capillary column (J&W Scientific, Palo Alto, CA). The initial column temperature was held at 60 °C for 1 min, ramped to 200 at 25 °C/min, ramped to 300 at 3 °C/min, and held for 10 min. Injection of 1 μL sample was conducted with an automatic sampler in the splitless

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