Size-dependent distribution and inhalation cancer risk of particle-bound polycyclic aromatic hydrocarbons at a typical e-waste recycling and an urban site

CrossMark

Pei Luo [a, d], Lian-Jun Bao [a], Shao-Meng Li [c], Eddy Y. Zeng [a, b, *]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] School of Environment, Jinan University, Guangzhou 510632, China
[c] Air Quality Research Division, Environment Canada, 4905 Dufferin Street, Toronto, Ontario M3H 5T4, Canada
[d] University of Chinese Academy of Sciences, Beijing 100049, China

ARTICLE INFO

ABSTRACT

Atmospheric particle size distribution of polycyclic aromatic hydrocarbons (PAHs) in a typical e-waste recycling zone and an urban site (Guangzhou) in southern China featured a unimodal peak in 0.56 −1.8 μm for 4−6 ring PAHs but no obvious peak for 2−3 ring PAHs at both sites. The atmospheric deposition fluxes of PAHs were estimated at 5.4 ± 2.3 μg m^{-2} d^{-1} in the e-waste recycling zone and 3.1 ± 0.6 μg m^{-2} d^{-1} in Guangzhou. In addition, dry and wet deposition fluxes of PAHs were dominated by coarse ($D_p > 1.8$ μm) and fine particles ($D_p < 1.8$ μm), respectively. Fine particles predominated the deposition of PAHs in the lung. The results estimated by incremental inhalation cancer risk suggested that particle-bound PAHs posed serious threat to human health within the e-waste recycling zone and Guangzhou.

1. Introduction

Air pollution has become a significant environmental and health concern in China (Chan and Yao, 2008; Chen et al., 2013). It was estimated that the life expectancy has been approximately 5.5 years lower for residents of northern China than for those living in southern China, likely because of sustained exposure to higher atmospheric total suspended particulates (Chen et al., 2013). In addition, atmospheric particulate matter has been shown to associate with abundant toxic pollutants, with polycyclic aromatic hydrocarbons (PAHs) accounting for the largest portion of the lung cancer risks (Hemminki and Pershagen, 1994). To demonstrate the severity of the situation, Zhang et al. (2009) estimated that incremental annual lung cancer risk due to inhalation exposure to PAHs was up to 6.5 per million people in China. Furthermore, the annual emission of PAHs in China (114 Gg y^{-1}) accounted for 22% of the total global emission in 2004 (Zhang and Tao, 2009), which is largely dissipated in the atmosphere initially.

Particle size distribution is a critical parameter for assessing health risks via inhalation, which has been shown to be size-dependent (Kameda et al., 2005; Luo et al., 2014a; Zhang et al., 2012c). Therefore, it is essential to consider the relative contributions of different sized particles to inhalation cancer risk by particle-bound PAHs in the atmosphere. In recent years, a number of studies on the size distribution of particle-bound PAHs have been conducted (Allen et al., 1996; Bi et al., 2005). However, the health risk of exposure to size-fractionated particle-bound PAHs is less well understood, especially in urbanized densely populated areas and primitive e-waste recycling zones. In addition, atmospheric dry and wet deposition is an important pathway for pollutants to cross the air-earth interface and to be removed from the atmosphere, which implicates greatly for the long-range transport potential of pollutants (Bidleman, 1988). Dry and wet deposition fluxes of particle-bound pollutants were also particle size-dependent (Luo et al., 2014b; Zhang et al., 2012a, 2012d).

Nearly 50 million tons of e-waste are generated worldwide each year, and a large portion of it is shipped to China where it is processed often with primitive methods to recover useful materials (Schluep et al., 2009; Zhang et al., 2012b). A variety of toxic contaminants, including PAHs, are released to the surrounding environment during e-waste recycling (such as e-waste burning), and

* Corresponding author. School of Environment, Jinan University, Guangzhou 510632, China.
E-mail address: eddyzeng@gig.ac.cn (E.Y. Zeng).

unprotected workers and residents living near e-waste processing facilities are exposed to these pollutants (Wong et al., 2007; Zhang et al., 2012b). In our previous study (Luo et al., 2014a), resident exposure to halogenated flame retardants, widely used in household electronic appliances, within an e-waste recycling zone was found to be of low health risk. It was therefore hypothesized that other contaminants such as PAHs may have posed more serious threats to human health. A previous study found that the inhalation cancer risk of parent and halogenated PAHs in an e-waste recycling area was 740–1200 cases per million people (Wang et al., 2012). However, this previous study only characterized PAHs in total suspended particulates.

To verify whether PAHs posed substantial human health risk in e-waste recycling areas as compared to typical urban regions, we conducted a sampling campaign in a typical e-waste recycling zone and an urban site. The main objectives of the present study were to examine the size distribution of particle-bound PAHs and estimate the atmospheric deposition fluxes and cancer risk of PAHs in the e-waste recycling zone and urban site. The acquired information is expected to aid a better understanding of the fate and health risk of PAHs associated with e-waste recycling activities.

2. Materials and methods

2.1. Sample collection

Size-fractionated particle samples were collected directly with a Micro-Orifice Uniform Deposit Impactor (MOUDI; MSP Corporation, Shoreview, MN, USA) from an e-waste recycling site in Qingyuan and an urban site in Guangzhou, Guangdong Province of southern China (Fig. S1 of the Supplemental Material; "S" indicates tables and figures in the Supplemental Material afterwards), in September–November 2012. Particle samples were separated into 11 fractions: >18, 10–18, 5.6–10, 3.2–5.6, 1.8–3.2, 1.0–1.8, 0.56–1.0, 0.32–0.56, 0.18–0.32, 0.10–0.18, and 0.056–0.10 μm on glass microfiber filters (Whatman International, Maidstone, England). Sampling took place at the ground level (1.5 m) and two rooftop levels (5 and 20 m) in Qingyuan and at 1.5 m and a rooftop level (20 m) in Guangzhou. Overall, 8 sets of size-fractionated samples were collected at each height in the e-waste recycling zone, and 7 and 4 sets of samples were at 1.5 m and 20 m in Guangzhou, respectively. Because outdoor activities mainly occur on the ground where mechanical processes and dust resuspension are significant, the data acquired at 20 and 1.5 m of both sampling sites were used for estimating atmospheric deposition fluxes and conducting human risk assessment, respectively.

2.2. Sample analysis

The size-fractionated samples were spiked with surrogate standards (acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, perylene-d_{12}, benzo[g,h,i]perylene-d_{12}, and coronene-d_{12}) and ultrasonically extracted three times with a mixed solvent containing *n*-hexane, dichloromethane, and acetone (1:1:1 in volume). Water was removed from the extracts with anhydrous sodium sulfate. After being concentrated to 50 μL under nitrogen flow, the internal standards (fluoranthene-d_{10}, pyrene-d_{10}, and dibenzo[a,h]anthracene-d_{14}) were added before instrumental analysis.

The concentrations of PAHs were determined with gas chromatographic mass spectrometry (Shimadzu QP2010 Plus, Kyoto, Japan) using electron impact ionization. Chromatographic separation of PAHs was achieved with a 30 m (0.25 mm i.d. and 0.25 μm film thickness) TG-5MS column (Thermo Scientific; West Palm Beach, FL, USA). The oven temperature was programmed starting from 60 °C (held for 1 min), increased to 200 °C at a rate of

6 °C min^{-1} (held for 2 min), to 300 °C at 8 °C min^{-1} (held for 20 min). Mass spectra were scanned from 50 to 500 mass units in the full-scan mode.

2.3. Quality assurance and quality control

The recoveries of the surrogate standards acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, perylene-d_{12}, benzo[g,h,i]perylene-d_{12}, and coronene-d_{12} were 65 ± 13%, 79 ± 11%, 102 ± 16%, 104 ± 21%, and 101 ± 22% in field samples and 64 ± 12%, 75 ± 12%, 101 ± 12%, 97 ± 14%, and 95 ± 20% in all blank samples. Concentrations of PAHs in all field samples were corrected by the corresponding procedural blanks in the same batch, but not corrected for the surrogate standard recoveries. The lowest calibration concentration divided by the actual sample volume was used as the reporting limit for a target compound. In the present study with an average air sampling volume of 21.6 m^3 and a final extract volume of 50 μL, the reporting limit was 11.6 pg m^{-3} for all PAHs.

2.4. Estimation of atmospheric deposition fluxes

Dry and wet deposition fluxes (F_{dry} and F_{wet}, respectively) of particle-bound PAHs were calculated by (Luo et al., 2014b; Zhang et al., 2012d)

$$F_{dry} = \sum (C_i \times V_i) \tag{1}$$

$$F_{wet} = W_T \times \sum (C_i \times \eta_i) \times Q \tag{2}$$

where C_i is the concentration of PAH in a specific size fraction i; V_i and η_i are dry deposition velocity and removal efficiency of a specific size fraction; W_T is the wet washout ratio (a conservative value of 10^5 was chosen for all PAHs because the W_T data of PAHs span several orders of magnitude from 10^3 to 10^7 (Birgül et al., 2011; Guo et al., 2014; Offenberg and Baker, 2002)); and Q is the monthly precipitation amount. Because wet deposition flux was highly dependent on rain amount, an average value of the monthly precipitation amounts in the past ten years was selected, i.e., 55 mm in October and November for Qingyuan (http://www.gdqy.gov.cn and therein) and 179 mm in September for Guangzhou (http://data.gzstats.gov.cn/gzStat1/chaxun/njsj.jsp). The V_i and η_i data were directly adopted from Zhang et al. (2012d) and Radke et al. (1980) (Table S1). The sum of dry and wet deposition flux is defined as total deposition flux.

2.5. Health risk assessment

Size-fractionated particles can be divided into three fractions in terms of their efficiencies of entering into the regions of the human respiratory system, i.e., inhalable (inhaled through the nose and/or mouth), thoracic (penetrating progressively into the lung), and respirable (reaching beyond the gas exchange region) fractions, based on the criteria given by the International Standards Organization and American Conference of Governmental Industrial Hygienists (Hinds, 1999). Particles can deposit in different regions of the human respiratory tract. The fractions of PAHs deposited in the head airway, tracheobronchial, and alveolar regions were estimated with the simplified equations from the International Commission on Radiological Protection (ICRP) model (Hinds, 1999; International Commission on Radiological Protection, 1994). More details about the models are provided in the Supplemental Material.

Human incremental lifetime cancer risk (*CR*) was estimated by