



Cumulative health risk assessment of halogenated and parent polycyclic aromatic hydrocarbons associated with particulate matters in urban air



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ABSTRACT

Halogenated polycyclic aromatic hydrocarbons (HPAHs) have been reported to occur widely in urban air. Nevertheless, knowledge about the human health risk associated with inhalation exposure to HPAHs is scarce so far. In the present study, nine HPAHs and 16 PAHs were determined in atmospheric particulate matter (PM) collected from Shenzhen, China to address this issue. Concentrations of Σ_9 HPAHs varied from 0.1 to 1.5 ng/m³ and from 0.09 to 0.4 ng/m³ in PM₁₀ and PM_{2.5} samples, respectively. As for individuals, 9-bromoanthracene, 7-bromobenz(a)anthracene, and 9,10-dibromoanthracene were the dominant congeners. Levels of Σ_{16} PAHs in PM₁₀ and PM_{2.5} samples ranged from 3.2 to 81 ng/m³ and from 2.8 to 85 ng/m³, respectively. Among individual PAHs, chrysene, benzo[b]fluoranthene, and indeno[1,2,3-c,d]pyrene were the main congeners. According to the season, concentrations of HPAHs and PAHs in atmospheric PM₁₀/PM_{2.5} samples show a similar decreasing trend with an order: winter > autumn > spring > summer. The daily intake (DI) of PM₁₀/PM_{2.5}-bound HPAHs and PAHs were estimated. Our results indicated that children have the highest DI levels via inhalation exposure. The incremental lifetime cancer risk (ILCR) induced by PM₁₀/PM_{2.5}-bound HPAHs and PAHs were calculated. The ILCR values showed a similar decreasing trend with an order: adults > children > seniors > adolescent. Overall, the ILCR values induced by HPAHs and PAHs were far below the priority risk level (10⁻⁴), indicating no obvious cancer risk. To our knowledge, this is the first study to investigate the human health risk associated with inhalation exposure to PM₁₀/PM_{2.5}-bound HPAHs.

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1. Introduction

Atmospheric particulate matter (PM) has been an environmental and health concern for decades, especially for coarse (PM₁₀, with an aerodynamic diameter > 2.5 and ≤ 10 μm) and fine (PM_{2.5}, with an aerodynamic diameter ≤ 2.5 μm) particles (Li et al., 2013). PM can cause a variety of urban air pollution and public health issues, such as haze episodes, visibility reduction, adverse health effects, and so on. Particle size is one of the important factors determining the adverse impacts of PM (Lagudu et al., 2011). For instance, PM₁₀ deposit primarily in the upper respiratory tract once inhaled, whereas PM_{2.5} can deposit into the tracheobronchial and even penetrate into the alveolar regions (Lin et al., 2005; Cao et al., 2014). Generally, the smallest particles pose the highest human health risks. In addition, PM₁₀ and PM_{2.5} have strong potential for adsorbing environmental toxicants because of

their large specific surface area (Betha et al., 2014). These adsorbed toxicants can exert influence on the respiratory and cardiovascular systems once they enter the human body through inhalation. So far, epidemiological studies have linked exposure to atmospheric PM to a wide range of adverse health outcomes, such as allergies, respiratory and cardiovascular diseases, and premature mortality (Zanobetti and Schwartz, 2009; Lagudu et al., 2011; Cheung et al., 2012; Bozlaker et al., 2014).

Polycyclic aromatic hydrocarbons (PAHs) are a group of ubiquitous environmental contaminants mainly coming from the incomplete combustion of organic materials (Qin et al., 2013; Zheng et al., 2014). Due to their potential toxic, mutagenic, and carcinogenic properties, PAHs are an important matter of concern for public health (Pieterse et al., 2013). Halogenated PAHs (HPAHs) are compounds with one or more halogen atom substituent attached to the aromatic rings of parent PAHs, including chlorinated PAHs (ClPAHs) and brominated PAHs (BrPAHs) (Sun et al., 2013). Compared with PAHs, knowledge about the occurrence of HPAHs in the environment is scarce (Kitazawa et al., 2006; Horii et al., 2008; Ohura et al., 2009; Sun et al., 2011; Ni and Zeng, 2012; Ma et al.,

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2013). Existing knowledge about the atmospheric HPAHs mainly focus on its occurrences, seasonal levels, and annual trends in the urban air of Shizuoka, Japan (Ohura et al., 2005, 2008b, 2009; Kitazawa et al., 2006). Only two studies regarding the occurrences of atmospheric HPAHs in China have been published (Wang et al., 2012; Ma et al., 2013). Meanwhile, most researchers focused on the occurrences of total suspended particulate (TSP)-associated HPAHs. Knowledge about the levels of PM₁₀/PM_{2.5}-bound HPAHs is limited (Ma et al., 2013). Specifically, no information about the occurrence of PM₁₀/PM_{2.5}-bound BrPAHs is available.

Existing knowledge about the human health risk of exposure to HPAHs is limited. Recently, the human health risk associated with dietary exposure to HPAHs has been evaluated for Chinese people (Ding et al., 2012, 2013; Ni and Guo, 2013). Nevertheless, studies about the human health risk of HPAHs through inhalation exposure is scarce. Only one study investigating the inhalation cancer risk of TSP-associated HPAHs has been published (Wang et al., 2012). So far, the human health risk associated with inhalation exposure to PM₁₀/PM_{2.5}-bound HPAHs remain unclear. The main objectives of the present study were to (1) measure the concentrations of HPAHs and PAHs in atmospheric PM₁₀/PM_{2.5} samples collected from Shenzhen, China; (2) estimate the daily intake of PM₁₀/PM_{2.5}-bound HPAHs and PAHs via inhalation exposure; (3) evaluate the potential health risk of PM₁₀/PM_{2.5}-bound HPAHs and PAHs.

2. Materials and methods

2.1. Sampling

Atmospheric samples of PM₁₀ and PM_{2.5} were collected at a rooftop location (20 m above ground level to avoid airflow obstruction) at the campus of Shenzhen Graduate School, Peking University (latitude 22°35'41.6" N, longitude 113°58'10.1" E) situated ca. 1 km from the nearest busy road and ca. 10 km from the city center. Samples were collected for 24 h for each of seven consecutive days for every month between September 2012 and August 2013. Atmospheric PM₁₀ and PM_{2.5} samples were collected on glass fiber filters (GFF; 20.3 cm × 25.4 cm, 0.3 μm nominal pore size, Whatman, Maidstone, England) using two high-volume air samplers (operating at a constant flow rate of ca. 1.05 m³/min) equipped with cutting heads for 10 μm and 2.5 μm particle sizes, respectively. Prior to sampling, GFFs were baked at 450 °C for 6 h in muffle furnace. After sampling, loaded GFFs were wrapped with prebaked aluminum foils and sealed with double layers of polyethylene bags. In total, 168 samples were collected. All of the samples were transported immediately to the laboratory and stored at –20 °C until extraction.

2.2. Chemicals

Nine individual HPAHs were determined in the present study. 9-chlorophenanthrene (9-ClPhe), 2-chloroanthracene (2-ClAnt), and 9,10-dichloroanthracene (9,10-Cl₂Ant) were purchased from Aldrich (St. Louis, MO). 1-bromopyrene (1-BrPyr), 2-bromofluorene (2-BrFle), 9-bromophenanthrene (9-BrPhe), 9-bromoanthracene (9-BrAnt), and 9,10-dibromoanthracene (9,10-Br₂Ant) were obtained from Acros Organics (Geel, Belgium). 7-bromobenz(a)anthracene (7-BrBaA) was purchased from Tokyo Kasei Kogyo Co., Ltd. (Tokyo, Japan). In addition, a standard solution of the 16 U. S. Environmental Protection Agency priority PAHs of the highest purity available was purchased from Chem Service, Inc. (West Chester, PA), including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fle), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene

(BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-c,d]pyrene (InP), dibenzo[a,h]anthracene (DBA), and benzo[g,h,i]perylene (BghiP). Internal standards (2-fluorobiphenyl and p-terphenyl-d₁₄) and surrogate standards (naphthalene-d₈, acenaphthylene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂) were purchased from Dr Ehrenstorfer GmbH (Augsburg, Germany). Anhydrous sodium sulfate was baked for 5 h in muffle furnace at 450 °C prior to use. Neutral silica gel (80–100 mesh) and alumina (80–100 mesh) were Soxhlet-extracted with methanol for 24 h and then with dichloromethane for another 24 h. Extracted silica gel and alumina were activated for 12 h at 180 °C and 250 °C, respectively, and then deactivated with distilled water (3%, w:w) prior to use. All glassware was hand-washed with detergent and tap water, rinsed with deionized water, and baked at 450 °C for at least 4 h prior to use. In addition, all organic solvents used were redistilled using a glass system.

2.3. Extraction and purification

Loaded GFFs were cut into pieces with surgical scissors, spiked with surrogate standards (naphthalene-d₈, acenaphthylene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂), and extracted with a mixture of 200 mL acetone: hexane (1:1, v:v) for 24 h. The extracts were concentrated to 2 mL using a rotary evaporator and then purified and fractionated using a glass column packed with alumina and silica gel (6:12, v:v) and anhydrous sodium sulfate (3 g). The first fraction of 6 mL of hexane was discarded. The second fraction, which contains most of the target compounds, was eluted with a mixture of 70 mL hexane and dichloromethane (7:3, v:v). This eluent was concentrated by rotary evaporation and further reduced with a gentle N₂ stream to a final volume of 500 μL. A known amount of internal standard (2-fluorobiphenyl and p-terphenyl-d₁₄) was added to the extracts prior to instrumental analysis.

2.4. Instrumental analysis

Concentrations of target analytes were determined using gas chromatography and mass spectrometry (GC–MS, Agilent 7890A GC equipped with 5975C MSD; Agilent Technologies, Foster City, CA) with a splitless injection. Gas chromatographic separation was accomplished using a 30 m DB-5MS fused silica capillary column (0.25 mm i.d., and 0.25 μm film thickness; J&W Scientific, Folsom, CA). High purity helium was used as the carrier gas. The mass selective detector was operated in the selected ion monitoring. The column oven temperature was initially programmed from 60 °C to 200 °C at 10 °C/min, from 200 °C to 214 °C at 2 °C/min, then from 214 °C to 254 °C at 5 °C/min (held for 2 min), and finally from 254 °C to 290 °C at 18 °C/min (held for 17 min).

2.5. Quality assurance and quality control

Field blanks (GFFs placed directly next to the air sampler throughout the sampling period) were collected each month. Procedural blanks and spiked blanks samples were processed with each batch of 10 samples to monitor procedural contamination. Recoveries of the surrogate standards spiked into cockroach samples were 68 ± 17%, 95 ± 8%, 86 ± 11%, 107 ± 13%, and 91 ± 10% for naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂, respectively. The quality control standards for HPAHs and PAHs were analyzed for each 10 samples to monitor the instrumental stability. Concentrations of the target analytes were determined with an internal calibration curve. The lowest concentration level from the calibration curve was defined as the reporting limit. Half of the reporting limit was used to

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