



PM_{2.5}-bound phthalates in indoor and outdoor air in Beijing: Seasonal distributions and human exposure via inhalation[☆]

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ABSTRACT

Phthalates (phthalates esters, PAEs) are ubiquitous contaminants in various indoor and outdoor environment. Exposure to PAEs exerts adverse effects on human health. Seasonal variations of air phthalate concentrations and paired indoor and outdoor air phthalate level are rarely known. In this study, six priority phthalates in PM_{2.5} were investigated in three indoor sites (a students' dormitory, a residential apartment and an office) and one outdoor site in Beijing, China across four seasons. PM_{2.5} samples were collected at indoor and outdoor environment simultaneously. Total PAEs in four sites were 468 ng/m³ (range: 9.52–1460 ng/m³), 498 ng/m³ (range: 11.2–4790 ng/m³), 280 ng/m³ (range: 4.08–1060 ng/m³), and 125 ng/m³ (range: 4.10–4000 ng/m³), respectively. DBP and DEHP were the most abundant PAEs across the four sampling sites, accounting for 76.3%–97.7% of the total PM_{2.5}-bound PAEs. Obvious seasonal variation of total PAEs was observed. PAEs concentrations were weakly or poorly correlated with PM_{2.5} levels. Indoor DBP and DEHP concentrations were much higher than those of outdoor, suggesting the importance of indoor DBP and DEHP sources. Principal component analysis revealed that cosmetics and personal care products, plasticizer and PVC products may be important sources for indoor PM_{2.5}-bound PAEs. Daily intakes of PAEs via inhalation for infants, student, and office-workers were 5.0, 0.8 and 0.9 μg/(kg-bw·day), respectively according to human exposure estimation.

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1. Introduction

Phthalates (phthalates esters, PAEs) have been used worldwide in industrial and consumer products as plasticizers in polyvinyl chloride (PVC) and other polymers, or as solvent or carriers in personal care products, varnishes, and coatings. The use of these products could gradually release PAEs into the environment, as the PAEs are not chemically bound to the products (Zota et al., 2014; Gong et al., 2014). Humans are exposure to PAEs through ingestion, inhalation and dermal pathways (Guo et al., 2014). Certain PAEs are highly concerned for its reproductive and developmental toxicity (Lyche et al., 2009; Bloom et al., 2015). Several studies have suggested a connection between PAEs exposure and adverse health outcomes, including reproductive system malformations, development abnormalities and cardiovascular diseases (Lyche et al.,

2009; Mariana et al., 2016; Katsikantami et al., 2016; Buck Louis et al., 2014). Di (2-ethylhexyl) phthalate, one of the most widespread phthalate plasticizer, has been classified as a probable human carcinogen (Group B2) by US EPA, and also has been listed as suspected carcinogen (1B) by European Union.

Along with the urbanization and modernization processes in China, large quantities of products containing PAEs are widely used in indoor environment, such as PVC flooring, building materials, cosmetics, children's toys, food packaging, etc. China is one of the world's largest consumers of phthalates and consumes a quarter of the total amount of PAEs in the world (Wang et al., 2010; Wang et al., 2017). Consequently, extensive use of PAEs brings adverse effects to public health in China. PAEs in the air, dust, soil, food, water, skin surface, personal care products have been investigated recently (Zhang et al., 2014; Bu et al., 2016; Wang et al., 2014; Song et al., 2015; Ji et al., 2014; Chen et al., 2012; Kang et al., 2012; Gong et al., 2014, 2016; Guo and Kannan, 2011; Guo et al., 2012, 2014; Wang et al., 2017), and phthalate metabolites in urines were also analyzed (Gao et al., 2016; Guo et al., 2011; Gong et al., 2015; Rocha et al., 2017). Exposure assessment among China showed that

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inhalation is an important pathway for phthalate intake. For infants, inhalation route accounts for 24.3%–96.8%, 2.1%–30.0% of the total daily exposure to DMP and DEHP, respectively (Bu et al., 2016; Wang et al., 2014; Wang et al., 2017; Guo and Kannan, 2011). Previous studies indicated that indoor phthalate concentrations were relatively high in China, compared to that in developed countries. Indoor air DBP and DEHP mean concentrations were 1650 ng/m³, 1510 ng/m³ in Xi'an (Wang et al., 2014) and 1099 ng/m³, 1125 ng/m³ in Hangzhou (Song et al., 2015), China, respectively, while those value were 112 ng/m³, 85.3 ng/m³ in Albany, New York, USA (Tran et al., 2017) and 99.9 ng/m³, 41.5 ng/m³ in Paris, France (Blanchard et al., 2014), respectively. Extensive indoor use of products containing phthalates could result in the high level of indoor air phthalate concentrations in China. Another possible reason is the relative high indoor particle concentration (Cao et al., 2012; Han et al., 2016; Zhao et al., 2015). Both experimental study and theoretical analysis showed that particles could enhance semivolatile organic compounds (SVOC) emission (Liu et al., 2012; Benning et al., 2013; Xu and Little, 2006). Benning et al. (2013) reported experimentally that particle concentration increasing from 100 to 245 µg/m³ could enhance DEHP emission from a vinyl flooring by a factor of 3–8.

Some measurements have been carried out to analyze indoor or outdoor airborne phthalate concentrations in China, however, little information is available on seasonal variations of phthalate concentrations both in indoor and outdoor environment. Previous studies on seasonal variations of indoor phthalate mainly focus on winter and summer period (Huo et al., 2016), and results about indoor phthalate levels across four seasons have not been reported yet.

The objective of this study was to improve our understanding of the seasonal variations of PAEs concentrations in air, the relationship of indoor and outdoor PAEs as well as their sources. Six priority PAEs in PM_{2.5} were determined in three indoor sites and one outdoor site in Beijing, China across four seasons, and the PM_{2.5} samples were collected at indoor and outdoor air simultaneously. Human exposure to PAEs via inhalation among different groups were also estimated. Beijing, the capital of China, experiencing serious PM_{2.5} air pollution, would also provide a chance to explore air phthalate levels at high particle levels.

2. Materials and methods

2.1. Site and sampling

Samples were collected in four sites in the urban area of Beijing, which included three indoor sites (a students' dormitory, a residential apartment and an office) and an outdoor site (Fig. S1, Supplementary Materials (SM)). The details of the four sampling dwellings and the participants were described in SM. All the indoor sites have been furnished for more than five years. Sampling was conducted from December 2014 to February, 2016 and included three synchronous sampling periods, described as synchronous sampling period I, II and III and shown in Table S1 (SM).

Filter-based PM_{2.5} samples from each sampling site were collected with two parallel Minivol Portable Air Samplers (Air-metrics, Springfield, Oregon, USA) on a Whatman 47 mm quartz filter and a Whatman 47 mm Teflon filter. All samplers were operated at a flow rate of 5 L/min and lasted for 72 h for each sample. Before sampling, the quartz filters were pre-heated at 500 °C for 4 h to remove any volatile components. After sampling, each loaded filter was stored in a refrigerator below 0 °C before gravimetric and chemical analysis.

2.2. Analytical method

2.2.1. Gravimetric analysis

Before and after sampling, Teflon filters were equilibrated for 24 h in a controlled chamber with the temperature of 20±1 °C and relative humidity of 50 ± 5%, and then weighed using an electronic balance (CPA-26P, Sartorius, German).

2.2.2. Composition analysis

The target compounds analyzed consisted of six PAEs are: dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DBP), benzyl butyl phthalate (BBP), di (2-ethylhexyl) phthalate (DEHP) and di-n-octyl phthalate (DNOP). The six PAEs of samples collected on the quartz filters were analyzed using a gas chromatographic mass spectrometry (GC-MS) (Trace GC Polaris Q, Thermo Scientific, Waltham, MA, USA), which operated in a selected ion monitoring mode. Detailed information about sample preparation as well as instrumental analysis are described in the SM.

2.2.3. Quality control and quality assurance

Following US EPA Method TO-13A (U.S. EPA, 1999), the lab blanks, standards and species-dependent recovery efficiencies were analyzed. PAEs recovery was evaluated by injecting standard mixtures including 6 PAEs formulated to mimic real sample concentrations into a blank quartz filter; the filter was then subjected to the entire analytical procedure. The mean recoveries of DMP, DEP, DBP, BBP, DEHP and DNOP were 97.0 ± 7.4%, 103.9 ± 11.2%, 108.8 ± 10.6%, 108.6 ± 12.6%, 118.3 ± 15.3% and 124.2 ± 13.8%, respectively.

3. Results and discussion

3.1. Characteristics of indoor/outdoor PAEs in PM_{2.5}

3.1.1. General characteristics of indoor/outdoor PAEs in PM_{2.5}

Fig. 1 shows the trend of the concentrations of PM_{2.5} and total PAEs in PM_{2.5} in indoor and outdoor sites during the whole sampling period. The average concentrations of PM_{2.5} for entire sampling period in the dormitory, office, residential home and outdoor were 52.1 µg/m³ (range: 11.2–176 µg/m³), 55.3 µg/m³ (range: 18.0–169 µg/m³), 35.5 µg/m³ (range: 5.41–149 µg/m³) and 68.5 µg/m³ (range: 13.6–266 µg/m³), respectively. Daily PM_{2.5} concentration exceeded the China National Ambient Air Quality Standards (75 µg/m³, level 2) (GB3095-2012) on 23.3% days in the dormitory, 22.7% days in the office, 9.2% days in the residential home and 63.3% days in the outdoor, respectively. The average concentrations of PM_{2.5} for the entire sampling period in four sites were all above the annual average of 35 µg/m³ (level 2), according to the National Ambient Air Quality Standards (GB3095-2012). The corresponding average concentrations of total PAEs in four sites were 468 ng/m³ (range: 9.52–1460 ng/m³), 280 ng/m³ (range: 4.08–1060 ng/m³), 498 ng/m³ (range: 11.2–4790 ng/m³) and 125 ng/m³ (range: 4.10–4000 ng/m³), respectively. As illustrated in Fig. 1, the concentrations of PM_{2.5} and total PAEs in four sites varied significantly during the entire sampling period. The PM_{2.5} concentration during the most polluted period (outdoor PM_{2.5} concentration reached 266 µg/m³ between Feb. 19th and 22nd, 2015) was almost 50 times higher than that during the cleanest period (outdoor PM_{2.5} concentration was 5.41 µg/m³ between May 30th and 2nd, 2015). While the total PAEs concentration varied about three scales of magnitude. In general, the correlation between ΣPAEs and PM_{2.5} in outdoor air was statistically positive but weak, with a Pearson correlation coefficient (ρ) of 0.33 (p < 0.01). Among six phthalate compounds in outdoor PM_{2.5}, DMP, DEP, DEHP (ρ ranging from 0.28

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