Atmospheric Environment 87 (2014) 146–152

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Indoor phthalate concentration and exposure in residential and office buildings in Xi'an, China

Xinke Wang [a,*], Wei Tao [a,b], Ying Xu [c], Jiangtao Feng [d], Fenghao Wang [a]

[a] School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, Xi'an 710049, Shaanxi Province, PR China
[b] China JK Institute of Engineering Investigation and Design, Xi'an 710043, Shaanxi Province, PR China
[c] Department of Civil, Architectural, and Environmental Engineering, The University of Texas at Austin, Austin, TX 78712, USA
[d] School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an 710049, Shaanxi Province, PR China

HIGHLIGHTS

- We measured indoor phthalates in air and dust from 28 buildings in Xi'an, China.
- Di-n-butyl phthalate (DnBP) and di(2-ethylhexyl) phthalate (DEHP) are the most abundant phthalates indoors in Xi'an.
- Correlation analysis shows indoor diisobutyl phthalate (DiBP), DnBP and DEHP might come from the same sources.
- The daily exposure to indoor phthalates in Xi'an was estimated.

ARTICLE INFO

Article history:
Received 4 October 2013
Received in revised form
20 December 2013
Accepted 6 January 2014
Available online 20 January 2014

Keywords:
Indoor air
Phthalate
Exposure
Field study

ABSTRACT

Indoor phthalate levels were investigated in 28 buildings, including 14 office and 14 residential buildings in Xi'an, China. Phthalate esters in the gas-, particle-, and dust- phase were measured separately. Four phthalates including dimethyl phthalate (DMP), diisobutyl phthalate (DiBP), di-n-butyl phthalate (DnBP) and di(2-ethylhexyl) phthalate (DEHP) were detected. The detection frequency of DnBP and DEHP was more than 90%. The concentrations of total phthalate esters ranged from 0.20 to 8.29 μg m^{-3} for the gas-phase, from 0.09 to 14.77 μg m^{-3} for the particle- phase and from 123 to 9504 μg g^{-1} for the dust- phase. The individual phthalate with the highest concentrations of 6.17 μg m^{-3}, 7.97 μg m^{-3} and 7228 μg g^{-1} respectively for gas-, particle- and dust- phase in all investigated rooms is all DiBP. The median concentration of the gas- and particle-phase DiBP (0.52 and 0.72 μg m^{-3}) and dust-phase DEHP (582 μg g^{-1}) were the highest. It was also found that the average concentrations of individual phthalates in residential buildings were often higher than in office buildings, and correlation analysis indicated that DiBP, DnBP and DEHP might come from the same sources. Based on the gas- and particle-phase concentrations measured, the particle-air partition coefficients of phthalates were estimated, and their logarithm values were found to be linearly correlated with the logarithm values of their octanol-air partition coefficients. Finally, the total daily exposure to indoor phthalates in air and dust was calculated, and ranged from 2.6 μg kg^{-1} day^{-1} (for adults) to 7.4 μg kg^{-1} day^{-1} (for toddlers).

1. Introduction

In recent years, phthalate esters (PAEs) have been identified by research as an indoor air pollutant that may have potential negative health consequences. Commonly, indoor phthalates are widely used as plasticizers in polyvinyl chloride (PVC) products such as flooring materials, toys, electronic cables or other plastic products and also exist in personal care products as solvents or carriers. About 3.5 million tons of phthalate products are consumed globally (Cadogan and Howick, 1996). When these products are used indoors, the phthalates are released, as they are not chemically bound to the polymer matrix or other materials. Therefore, phthalates are ubiquitous in indoor environments.

The presence of phthalates indoors has been correlated with some adverse health effects. As typical endocrine-disrupting chemicals, exposure to phthalates may result in reproductive and developmental problems. Studies have shown that some phthalates are linked with problems in male reproductive development such as poor semen quality (Liu et al., 2012; Swan, 2008) and decreased

* Corresponding author. Tel.: +86 29 83395127; fax: +86 29 83395100.
E-mail addresses: wangxinke@mail.xjtu.edu.cn, wangxinke98@gmail.com (X. Wang).

1352-2310/$ – see front matter © 2014 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.atmosenv.2014.01.018

anogenital distance (Swan et al., 2005), and also to problems in female fertility and development including pregnancy loss (Toft et al., 2012), shortened pregnancy duration (Latini et al., 2003), pre-mature breast development and precocious puberty (Wolff et al., 2010). Epidemiologic investigations have also revealed an association between exposure to indoor phthalates and childhood health. Numerous studies indicate that there are strong associations between phthalates and children's asthma and allergies (Bornehag et al., 2004; Jaakkola and Knight, 2008). In addition, some research suggests a relationship between phthalate exposure and infant neurodevelopment (Kim et al., 2011).

Phthalates are semi-volatile organic compounds (SVOCs) and are especially easily adsorbed on indoor particular matters and surfaces due to low vapor pressure (Weschler and Nazaroff, 2010). The gas-, particle-, surface- and dust- phases of phthalates play important roles in the migration of indoor phthalates from their sources to human bodies through three main routes, which include inhalation, dermal adsorption, and dietary uptake (Weschler and Nazaroff, 2012). An investigation of indoor phthalates concentrations in different phases is the basis for an estimation of phthalate exposures. Indoor phthalate levels have been measured in several countries, such as China (Guo and Kannan, 2011; Lin et al., 2009; Pei et al., 2013; Zhang et al., 2013), Denmark (Langer et al., 2010), Germany (Becker et al., 2009), Norway (Rakkestad et al., 2007), Sweden (Bergh et al., 2011; Bornehag et al., 2005) and U.S. (Guo and Kannan, 2011; Hwang et al., 2008; Rudel et al., 2003). The comparisons show that indoor phthalate levels vary sharply in different locations. Because about a quarter of the total amount of phthalates used in the world are produced in China (Wang et al., 2010), there is an increasing tendency to be concerned about indoor phthalate pollution in China. However, the data for indoor phthalate concentrations in the western region of China is still rare. In this study, several typical phthalate esters in their gas-, particle-, and dust-phases were measured in indoor environments in Xi'an, a representative city of western China, and phthalate exposures were estimated based on that data.

2. Methods

2.1. Chemicals

17 common phthalate esters are selected as target compounds in the investigation and their related information is listed in Supplementary Information (S2), which are selected according to the standard mixture of phthalate esters with 1000 μg mL^{-1} from Chemservice Inc.

2.2. Building information

This study involved a total of 28 buildings, consisting of 14 office buildings and 14 residential buildings. All the buildings had interior decoration of basic levels and only common indoor materials were used. In the buildings, the floor was covered by ceramic tiles or wooden floorboards and the walls were coated with latex paint, gypsum or wallpaper. All residential buildings were constructed less than 5 years previously and office buildings less than 10 years.

2.3. Sampling

Gas- and particle-phase phthalates were collected by an active SVOC sampler (the detailed information can be found in the Supporting Information S3) as designed according to U.S. EPA method TO13-A (EPA, 1999). The gas-phase phthalates were adsorbed by polyurethane foam (PUF with size Φ 90 mm × 50 mm) and the particles with phthalates were trapped on a glass-fiber filter with effective diameter 80 mm and aperture diameter less than 10 μm. The sampling flow rate was 100 L min^{-1} and the sampling duration was 24 h for one sampling point. Prior to the sampling, the sampling media was pre-conditioned to decrease phthalate ester residues. The PUFs were dipped into the boiled water and was twisted back and forth several times. The glass fiber membranes were baked at 400 °C in a Muffle furnace for over 4 h and then were weighed by an electric balance with 1 mg precision before and after sampling. Before sampling, the PUFs were extracted by methylene dichloride in a Soxhlet extractor and then kept at 4 °C as operation described in "Measurement" part after sampling.

Indoor dust from non-walking surfaces in the selected buildings was swept on to a glass-fiber membrane. Then the glass-fiber membrane together with the collected dust was quickly transferred in a sealed glass bottles. Prior to the instrumental analysis, all samples were stored at 4 °C.

The sampling was conducted from September 9th, 2012 to January 9th, 2013. When starting sampling, the indoor temperature was recorded and ranged from 14.6 °C to 21.5 °C in the period (Seen in Supplementary Information S4).

2.4. Measurement

After sampling, the samples were extracted separately in 6 cycles per hour for total 8 h by methylene dichloride through a Soxhlet extractor. Then the sample was concentrated into a 1 mL volume through a stream of clean nitrogen at 40 °C. The final concentrated solution was kept at 4 °C until GC–MS analysis.

Phthalate concentrations were determined by a gas chromatography–mass spectrometry instrument (Thermo Scientific ISQ) through splitless injection on a 30 m HP-5MS column (Agilent Scientific Inc.; 30 m × 0.25 mm[inner diameter] × 0.25 μm [film thickness]; the column flow rate: 0.5 mL min^{-1}) carried by helium gas. The temperature program of the oven was 70 °C for 4 min, risen at 10 °C min^{-1} to 280 °C and maintained for 4 min. The coupled mass selective detector was operated in electron impact (EI) mode using selected ion-monitoring (SIM).

2.5. Quality assurance and quality control

Before the investigation, two field blanks, two transportation blanks and two process blanks for each sampling point were analyzed together with the samples. And the concentrations of target PAEs were all lower than the LOD (see Supplementary Information S5). A recovery efficiency experiment was conducted ($n = 4$) using a 1 μg μL^{-1} standard mixture of 17 phthalate esters in hexane (Chemservice Inc.) added to the blank glass fiber membranes and PUF filters (10, 20, 30, 40 and 50 μL respectively). Through the same pretreatment and lab analysis procedure as the samples, the recovery efficiencies were obtained and ranged from 73% to 94%. The precision of the method was assessed by replicate experiments ($n = 6$) by adding a 1 μg standard mixture to a blank sampling medium. The relative standard deviation (RSD) was found to range from 7.5% to 19.7%.

2.6. Statistical analysis

IBM SPSS Statistics version 19.0 software was employed for data analysis. Mean, median, standard deviation, and ranges were given. Non-parametric independent-samples with the Mann–Whitney *U* test method were used to compare the difference between concentrations in homes and offices. Spearman's rank correlation was applied for analyzing correlations between phthalate concentrations in different phases.