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Levels of phthalate esters in settled house dust from urban dwellings with young children in Nanjing, China

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HIGHLIGHTS

► Phthalate esters (PEs) were measured in settled house dust from urban dwellings.
► Di-2-ethylhexyl phthalate and di-n-butyl phthalate are the most abundant.
► Solid-wood flooring wax and cosmetic and personal care products are two PE sources.
► Flooring material, dampness and humidifier use potentially influence PE levels.

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ABSTRACT

To investigate the levels and possible determinants of phthalate esters (PEs) in settled house dust from urban dwellings with young children, dust was collected from 215 urban houses in Nanjing, China, and 145 outdoor settled dust samples were collected nearby. Six PEs were measured by gas chromatography/mass spectrometry. All PEs were detected in the dust from approximately 90% of the houses, with the exception of dioctyl phthalate (DOP), which had only a 59% detection rate. Di-2-ethylhexyl phthalate (DEHP) and di-n-butyl phthalate (DBP) were the most abundant PEs, with geometric means of 110 and 16.4 $\mu g\ g^{-1}$, respectively, and maximal concentrations 9950 and 2150 $\mu g\ g^{-1}$. Factor analysis showed that DBP, DEHP and benzyl butyl phthalate (BBP) might come from the same source and were significantly influenced by the use of solid-wood floor wax. High BBP, DEHP, DOP and total PE levels were associated with indices of dampness, and high DOP was associated with humidifier use. In conclusion, six PEs are ubiquitous in urban settled house dust in Nanjing, China, and both plastic materials and cosmetic and personal care products are important sources. Flooring material, dampness and humidifier use potentially influence house dust PE levels.

1. Introduction

Concentrations of chemical contaminants, including semi-volatile organic compounds (SVOCs), such as polychlorinated biphenyls, flame retardants and pesticides, are often higher in indoor air than in outdoor air (Rudel and Perovich, 2009). Many indoor contaminants are absorbed by particulate matter that is initially suspended in the air and later settles as dust. Settled house dust is thus considered as a medium of exposure and a global indicator of residential contamination (Butte and Heinzow, 2002; Lioy et al., 2002; Weschler and Nazaroff, 2010). Chemical exposure through settled house dust is very important for infants and toddlers, who are at highest risk because they frequently place their hands in their mouth and ingest dust.

Phthalate ester (PE) levels in dust have been reported to be associated with asthma and allergies in children (Bornehag et al., 2004; Jaakkola et al., 2004; Kolarik et al., 2008b). Simultaneously, given that neurodevelopmental processes such as myelination are not completed until adolescence (Rice and Barone, 2000), and that inadvertent dust ingestion through hand-to-mouth play has been proven to be a major route of PE uptake in young children (Heudorf et al., 2007; Sathyanarayana, 2008; Wormuth et al., 2006), PE levels

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in settled house dust have recently become considered as particularly important for young children. Prenatal PE exposure has been linked to neurodevelopmental toxicity, and there is a negative association between such exposures and children's intelligence and behavior (Engel et al., 2010; Kim et al., 2011). A study from South Korea reported an inverse relationship between urinary concentrations of phthalate metabolites in elementary school students and their intellectual functioning (Cho et al., 2010).

Studies have reported PE concentrations in different localities and countries of the order of tens to thousands of micrograms per gram of settled house dust (Becker et al., 2009; Bornehag et al., 2005; Hwang et al., 2008; Langer et al., 2010; Rudel et al., 2003). In 2006, the total worldwide PE use was 6.65 million metric tons, one-quarter of which was in China (Tao and Liang, 2008). Despite the high use of PEs in China, only two studies with small sample sizes have reported the occurrence of PEs in settled house dust in that country, with a large difference between their findings (Guo and Kannan, 2011; Lin et al., 2009). The PE levels in settled house dust in China and the magnitude of childhood exposure are therefore unclear, and information on the possible determinants of PE concentrations is needed. In this study, PE concentrations in settled house dust were measured in Nanjing, China to evaluate the extent of PE pollution in urban dwellings with young children, and its possible determinants and sources.

2. Methods

2.1. Subject recruitment and dust sampling

The study was approved by the Institutional Ethics Committee of Nanjing Medical University and Institutional Review Board. All houses from which dust samples were collected and located in an urban district of Nanjing, China; subject recruitment is described elsewhere (Wang et al., 2011). All dust was sampled during March to June 2011, when domestic heating is not used. One sample of dust from each house was collected using commercial vacuum cleaners and paper bags on the floor surface in each dwelling where preschoolers commonly played; simultaneously, settled dust on an outdoor ground surface near the house was sampled with a handheld brush. For other details about the sampling process, please see the Supporting information. Two hundred and thirty samples of indoor dust and 150 of outdoor dust were collected. After excluding mislabeled, mis-stored, and insufficient dust samples, 215 samples of indoor dust and 145 paired outdoor samples were available for the final analysis.

2.2. Building investigations

During dust sampling, the investigators were asked to collect information on duration of residence, indoor decoration level (no decoration; simple decoration: <0.1 million Yuan; moderate or expensive decoration: ≥0.1 million Yuan), building materials (including floors, furniture and walls), floor level, frequency of natural ventilation through open windows (<1 h d^{-1}, 1–4 h d^{-1} or >4 h d^{-1}), frequency of vacuuming or cleaning (every 1–3 d, every 4–7 d, ≤14 d or >14 d), indicators of dampness (presence of condensation, mold growth, water stains or wet towels) and whether the residence was near a main road (<500 m from the child's home to a two- or more-lane motorway).

2.3. PEs in dust

Pretreatment and laboratory measurement of PEs in dust was conducted following the method described by Kanazawa et al. (2010), with modifications. Coarse material was removed from the raw dust and particles were separated from fibers by sieving through a mesh that was <150 μm in size. Acetone was added to each sieved dust sample and extraction was performed in an ultrasonic bath. The supernatant was concentrated and then acetone added to 0.1 mL for analysis by gas chromatography/mass spectrometry (TraceGC ultra-Trace DSQ; ThermoFisher Scientific, Waltham, MA). Other details for the measurements were described elsewhere (Lu et al., 2012) and the main points were included in the Supporting Information.

2.4. Quality assurance and quality control

Fifty milligrams of the house dust were ashed in a muffle furnace at 800 °C for 1 h to destroy all possible PEs (blank house dust without PEs). A recovery efficiency study was conducted ($n = 6$) using 100 ng of 6 PE standard mixtures added to the blank house dust. The pretreatment and lab analysis procedures were the same as the samples. The recovery efficiencies ranged from 88% to 111%. The precision of the method was evaluated by replicate analyses ($n = 6$) of blank house dust that contained 100 ng of 6 PE standard mixtures. The relative standard deviation (RSD) ranged from 6% to 19%. The average relative percentage difference (RPD) for the PEs ranged from 4.2% to 9.6%. The instrumental detection limit (IDL) was determined on the basis of the response at a signal-to-noise ratio (S/N) of 3. The method detection limits (MDL) under the present chromatographic conditions were calculated by the IDLs, the volume of extracts and the sample weights. The values of the IDLs, MDLs, recovery efficiencies and RSD are summarized in Table S1. Only glass centrifuge tubes, glass pipettes and glass syringes were used in the extraction and clean-up steps, and all glassware were cleaned by ultrasonication in acetone for over 30 min prior to their use. Standard calibration curves were prepared for each batch of 20 samples. Before the sample injection, the syringe was washed three times with dichloromethane, and then three times with n-hexane.

2.5. Statistical analysis

PE concentrations below the detection limit were replaced by half of the detection limit values. The Kolmogorov–Smirnov test showed that natural logarithm transformed (ln-transformed) PE concentrations were normally distributed as reported previously (Ott, 1990). Thus ln-transformed data were used in the subsequent statistical analysis. Geometric mean (GM) and geometric standard deviation (GSD) were used to describe the distribution of PE levels. Arithmetic mean, median, 95th percentile and range were also used to compare the data with those reported in the literature. Pearson's correlation was used for bivariate association. The paired-sample *t*-test was used to analyze the difference between indoor and outdoor dust samples. The Student's *t*-test was used to analyze the difference in PE concentrations between two subgroups of the categorized factors. One-way analysis of variance was used to check the difference in PE concentrations between subgroups of the categorized determinants and the least significant difference test was used for multiple comparisons. Exploratory factor analysis was conducted on the ln-transformed PE concentrations of settled house dust and outdoor dust to identify potential structures within the data. Factor designations were confirmed using proc factors with method = ML and Promax Rotation. All analyses were performed using SPSS 16.0 (SPSS Inc., Chicago, IL).

3. Results and discussion

3.1. PEs in settled house dust

Table 1 presents summary statistics for PE concentrations in settled house dust. The most frequently identified PEs were DEHP