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Indoor phthalate concentration in residential apartments in Chongqing, China: Implications for preschool children's exposure and risk assessment

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HIGHLIGHTS

- Phthalate levels in residences were studied for the first time in Chongqing.
- Di(2-ethylhexyl) phthalate and dimethyl phthalate were the most abundant.
- Preschool children's exposure to phthalates was estimated.
- Exposure to DnBP and DEHP may be harmful to preschool children.
- A detailed study on the exposure factors is needed.

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ABSTRACT

Six phthalates – dimethyl phthalate (DMP), diethyl phthalate (DEP), di(isobutyl) phthalate (DiBP), di(n-butyl) phthalate (DnBP), butyl benzyl phthalate (BBzP) and di(2-ethylhexyl) phthalate (DEHP) – in indoor gas-phase and dust samples were measured in thirty residential apartments for the first time in Chongqing, China. Monte-Carlo simulation was used to estimate preschool children's exposure via inhalation, non-dietary ingestion and dermal absorption based on gas-phase and dust concentrations. Risk assessment was evaluated by comparing the modeled exposure doses with child-specific benchmarks specified in California's Proposition 65. The detection frequency for all the targeted phthalates was more than 80% except for BBzP. DMP was the most predominant compound in the gas-phase (median = 0.91 μg/m^3 and 0.82 μg/m^3 in living rooms and bedrooms, respectively), and DEHP was the most predominant compound in the dust samples (median = 1543 μg/g and 1450 μg/g in living rooms and bedrooms, respectively). Correlation analysis suggests that indoor DiBP and DnBP might come from the same emission sources. The simulations showed that the median DEHP daily intake was 3.18–4.28 μg/day/kg-bw in all age groups, suggesting that it was the greatest of the targeted phthalates. The risk assessment indicated that the exposure doses of DnBP and DEHP exceeded the child-specific benchmarks in more than 90% of preschool children in Chongqing. Therefore, from a children's health perspective, efforts should focus on controlling indoor phthalate concentrations and exposures.

1. Introduction

Phthalates are a group of chemicals used during the manufacturing processes of a range of household and industrial products. Globally, more than 200 million kilograms of phthalates are produced every year (Gong et al., 2014). Phthalates are used widely as plasticizers in polyvinyl chloride (PVC) flooring, personal care products, children's toys, food packaging, and building materials because of their unique physical and chemical characteristics (Fromme et al., 2004; Kang et al., 2012). Higher molecular weight phthalates such as di(2-ethylhexyl) phthalate (DEHP) and butyl

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benzyl phthalate (BBzP) are often used in PVC production. Lower-molecular-weight phthalates, such as dimethyl phthalate (DMP) and diethyl phthalate (DEP) are mainly found in cosmetics, personal care products and surface coating materials (Abb et al., 2009; Kang et al., 2012). Di(isobutyl) phthalate (DiBP) and di(n-butyl) phthalate (DnBP) are commonly used in plastic and body care products (Gong et al., 2014).

Exposure to phthalates has been linked to significant adverse health effects. DiBP, DnBP, BBzP and DEHP have been reported to disrupt reproductive endocrine production in both experimental animals and humans (Barlow et al., 2004; Davis et al., 1994; Koch et al., 2012; Zacharewski et al., 1998). Reproductive health problems have been documented to have connections with exposure to phthalates, including decreased reproductive hormone levels in adult males, poor semen quality, reduced anogenital distance in male infants (Bornehag et al., 2015; Koch et al., 2011), pre-mature breast development and pregnancy loss (Toft et al., 2012; Wolff et al., 2010). Other studies have reported obvious correlations between asthma and allergic symptoms and exposure levels to phthalates in children's apartments (Bekö et al., 2015; Callesen et al., 2014; Whyatt et al., 2012). It has also been reported that phthalate exposure is a potential contributor to neurodevelopment problems and obesity (Hatch et al., 2010; Whyatt et al., 2012). On the other hand, DEHP acts as a rodent liver carcinogen via peroxisome proliferation (Carpenter et al., 1953). Moreover, the U.S. Environmental Protection Agency (EPA) has pointed out that DEHP may generate potential carcinogenic effects on animals and humans according to related carcinogenicity data from experiments and investigations. The Integrated Risk Information System (IRIS) has classified DEHP as a B2 substance, that is, a probable human carcinogen (EPA, 1997).

Phthalates are semi-volatile organic compounds (SVOCs). In comparison to general gaseous pollutants, e.g., formaldehyde and volatile organic compounds (VOCs), phthalates can be easily adsorbed into indoor airborne particulate matters and surfaces (Weschler and Nazaroff, 2010) due to their low vapor pressure. Since phthalates are not chemically bound to those products, they will be continuously leached and released into the indoor environment. Thus, the ubiquitous existence of phthalates in indoor dust, air, and even food has been reported in several studies (Bornehag et al., 2005; Gaspar et al., 2014; Kang et al., 2012; Pei et al., 2013). Human exposure pathways to phthalates include inhalation, oral ingestion and dermal absorption indoors (ATSDR, 2005). Phthalate metabolites have been reported in the human body fluids like urine and blood (Fromme et al., 2013; Silva et al., 2005; Toft et al., 2012). In comparison to adults, preschool children undergo physical growth and development, and are more susceptible to these chemicals indoors as a result of their exploratory behavior without fully-developed physical frame (Gaspar et al., 2014; Koch et al., 2011). Infants and children have higher exposures than adults because their intake is greater per unit body weight via multiple pathways. They tend to be more exposed due to their frequent skin contact with surfaces and mouthing of fingers or plastic toys (Bekö et al., 2013). Thus, in residential buildings, which are dominant places for preschool children's daily activities, chronic phthalates exposure may be an important burden to their health.

Therefore, more attention has been paid to monitoring indoor phthalate levels. Indoor air and settled dust are major concerns as they are considered to be typical indicators of residential contamination as well as media of human exposure (Lioy et al., 2002; Weschler and Nazaroff, 2010; Zhang et al., 2013). Numerous studies in Denmark, Sweden, Germany, the U.S. (Fromme et al., 2004; Guo and Kannan, 2011; Koch et al., 2011; Soeborg et al., 2012), and China (Guo and Kannan, 2011; Pei et al., 2013; Wang et al., 2014; Zhang et al., 2013) have reported phthalate levels in houses. Children's exposure to phthalates via multiple pathways was also studied by indirect estimation through daily intake calculation (Guo and Kannan, 2011; Kang et al., 2012; Pei et al., 2013; Wang et al., 2014) or combined with biomonitoring (Bekö et al., 2015, 2013; Callesen et al., 2014; Gong et al., 2015; Langer et al., 2014). From a risk assessment perspective, most investigations have directly compared exposure levels with common benchmarks, e.g. U.S. EPA's Reference Doses (RfD) (Kim et al., 2011), the European Chemical Safety's Derived No-Effect Level (DNEL) (Lee et al., 2014) and European Food Safety Authority's Tolerable Daily Intake (TDI) (Bekö et al., 2013; Koch et al., 2011; Soeborg et al., 2012), but have not given information on the differences in health risk categories. Moreover, sensitivity to phthalates exposure as a function of children's age has been seldom used in the selection of exposure benchmarks for risk estimates. So far, few investigations have reported phthalate levels and the corresponding health risks to children in residential apartments in China. Although Southwestern China is experiencing rapid modernization and urbanization, information on indoor phthalate exposure and risk assessment of preschool children is relatively scarce. Thus, a detailed study on phthalate levels as well as exposures via each pathway in residential apartments is necessary.

Chongqing is one of four municipalities achieved province status in China. Located in Southwestern China, its urbanization and industrialization during the last three decades has been rapid. Phthalate pollution in residential apartments is likely prevalent because PVC flooring is commonly used, so are plastic materials. The targets of this study were living rooms and children's bedrooms, which were regarded as being the locations where young children spend most of their time. The aims of this study were (1) to estimate and rank the phthalate exposure levels of preschool children via multiple pathways in residential apartments in Chongqing and (2) to assess child-specific reproductive and cancer risk separately among preschool children in Chongqing with the assistance of the Office of Environmental Health Hazard Assessment (OEHHA)'s Proposition 65.

2. Materials and methods

2.1. Sampling site and methods

Thirty residential apartments were selected in the following Chongqing downtown districts: Shapingba, Dadukou, Jiulongpo, Jiangbei, Nan'an and Yubei. Characteristics of the thirty volunteered residential apartments were as follows: (a) at least one year elapsed since the last renovation to avoid excessive concentration values from the tested results; and (b) located in areas with relatively higher population density to conform to the general living circumstances of rapidly developing area of China. The thirty sampled apartments represented Chongqing's typical urban residential environment. Detailed information on the downtown areas of Chongqing and sampling locations are depicted in Supplementary Information S1. The on-site investigation was carried out from November 14th, 2014 to February 2nd, 2015. During the survey time, a normal state of residential activities was maintained. Indoor temperature, relative humidity and atmospheric pressure were recorded.

Gas-phase samples in the targeted apartments were collected from living rooms and children's bedrooms using polyurethane foams (PUFs) as the absorption media. Each collection was done at a height corresponding to the children's breathing zone (0.6–1.0 m). Air was drawn at approximately 4 L per minute (LPM) through PUFs (with size $\Phi 25$ mm $\times$ 70 mm) during the sampling. PUFs were pre-cleaned by extraction in dichloromethane for about 48 h and dried