Environment International 96 (2016) 16–23

Contents lists available at ScienceDirect

Environment International

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

Exposure to phthalates in house dust and associated allergies in children aged 6–12 years

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A R T I C L E I N F O

Article history:
Received 7 May 2016
Received in revised form 25 August 2016
Accepted 25 August 2016
Available online xxxx

Keywords:
Allergy
Child
Daily intake
House dust
Phthalates
Urinary metabolites

A B S T R A C T

Phthalates are widely used as plasticizers in household products. Several studies have reported an association between phthalate exposure and an increased risk of allergies. The present study estimated phthalate exposure in children aged 6–12 years and assessed potential correlations with allergies. House dust samples were collected from floors and multi-surface objects >35 cm above the floor. Urine samples were collected from the first morning void of the day. Daily phthalate intake (DI_{dust} and DI) was estimated using both house dust and urinary metabolite concentrations. Exposure to di-2-ethylhexyl phthalate (DEHP) in floor dust was associated with parental-reported rhino-conjunctivitis. After stratification by gender, this trend was found to only occur in boys. Furthermore, urinary mono-isobutyl phthalate was inversely associated with parental-reported wheeze in boys. DI_{dust} of benzyl butyl phthalate (BBzP) and DEHP were significantly correlated with DI_BBzP and DI_DEHP, respectively. These correlations were stronger with floor than with multi-surface dust. Our results suggest that, among Japanese children, house dust from low surfaces, such as living room floors, might play a meaningful role in the indoor environmental exposure pathway for BBzP and DEHP.

1. Introduction

The adverse health effects of phthalates as endocrine disruptors are a growing public concern. Phthalates are widely used as plasticizers in household products, such as polyvinyl chloride (PVC) flooring and building materials. Although di(2-ethylhexyl) phthalate (DEHP) is regulated for use in toys and food packaging, and Japanese consumption and production of phthalates have decreased over the past 10 years (Bizzari et al., 2013), DEHP is still the most widely used phthalate, used mainly as a plasticizer in PVC. Several studies, in both animals and humans, have reported that phthalate exposure is associated with adverse health effects in children, including effects on reproductive, neurological, and pubertal development, and obesity (Bornehag and Nanberg, 2010; Lovekamp-Swan and Davis, 2003; Martino-Andrade and Chahoud, 2010). Furthermore, phthalates found in house dust are considered to increase the risk of allergies (Ait Bamai et al., 2014; Bornehag et al., 2004; Kolarik et al., 2008). Although the major routes of phthalate exposure are thought to be through diet and the use of personal care products (PCP) (Koch et al., 2013), house dust may also be important.

Animal studies have indicated that some phthalates may enhance allergic responses by acting as adjuvants for the production of cytokines and/or immunoglobulins responsible for allergic sensitization (Koike et al., 2010; Larsen et al., 2001a; Lee et al., 2004; Tanaka et al., 2013). In an epidemiological study, Beko et al. (2015) evaluated the association between allergies in children and the levels of both urinary phthalate metabolites and phthalates in house dust. They reported that allergic sensitization was significantly associated with certain house dust phthalates, but not with urinary phthalate metabolite levels (Beko et al., 2015). Other previous studies reported that the highest quartile of benzyl butyl phthalate (BBzP) metabolites in Taiwanese children aged 2 years old was associated with an increased risk of atopic dermatitis among children at 2 years, but not at 5 years of age (Wang et al., 2014). Similarly, the highest quartiles of di-iso-nonyl phthalate (DiNP) and di-iso-decyl phthalate metabolites increased the risk of current asthma in 10-year-old Norwegian children (Bertelsen et al., 2013).

Abbreviations: BBzP, benzyl butyl phthalate; DBP, dibutyl phthalate; DEHP, di-2-ethylhexyl phthalate; DEP, diethyl phthalate; DiBP, di-iso-butyl phthalate; DiNP, di-iso-nonyl phthalate; DMP, dimethyl phthalate; DnBP, di-n-butyl phthalate; MBzP, mono benzyl phthalate; MCNP, mono-(carboxynonyl) phthalate; MCPP, mono-(3-carboxypropyl) phthalate; MECPP, mono-(2-ethyl-5-carboxypentyl) phthalate; MEHP, mono-(2-ethylhexyl) phthalate; MEOHP, mono-(2-ethyl-5-oxohexyl) phthalate; MiBP, mono-isobutyl phthalate; MnBP, mono-n-butyl phthalate; PVC, polyvinyl chloride; SVOC, semi-volatile organic compounds.

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http://dx.doi.org/10.1016/j.envint.2016.08.025
0160-4120/

Gender-specific responses to phthalate exposure were considered by Ku et al. (2015), who reported that postnatal DEHP and diethyl phthalate (DEP) exposures were associated with asthma among Taiwanese boys at 8 years of age (Ku et al., 2015). However, data regarding the role of routes of phthalate exposure are still limited.

One way to assess indoor phthalate contamination epidemiologically is by measuring phthalate concentrations in house dust. Numerous studies have detected phthalates in house dust (Abb et al., 2009; Bornehag et al., 2005; Jaakkola et al., 1999; Langer et al., 2010; Oie et al., 1997), but reports on the association with allergies are limited. A Swedish study found that DEHP exposure via house dust was associated with asthma, and BBzP was associated with eczema and rhinitis in children aged 3–8 years old (Bornehag et al., 2004). Consistent results were reported for DEHP in Bulgarian children (Kolarik et al., 2008). Callesen et al. (2014b) did not find a significant association between phthalates in dust and diagnosed allergies among children in Denmark, but they did report a positive association between DEHP in dust and parental-reported wheeze. In most studies, dust samples were only collected from surfaces above floor level (Beko et al., 2015; Bornehag et al., 2004; Callesen et al., 2014b; Hsu et al., 2012; Kolarik et al., 2008). We previously reported that the concentration of phthalates in house dust collected from surfaces above floor level were higher than in dust collected from the floor (Ait Bamai et al., 2013). However, dose-response associations between self-reported allergies and phthalates, including DEHP with allergic conjunctivitis, both BBzP and di-iso-butyl phthalate (DiBP) with atopic dermatitis, and DiNP with allergic rhinitis, were only found in floor surface dust (Ait Bamai et al., 2014). Reported phthalate exposure levels, such as levels of DEHP in house dust, differ among Swedish, Bulgarian, Danish, and Japanese studies. House dust levels of DEHP in Sweden were low, and those in Denmark were very low; however, levels in Bulgaria were high, and those in Japan were very high (Ait Bamai et al., 2013). These studies had different designs, and in particular, studies differed in the type of buildings from which dust was collected for analysis. Our previous findings indicated that allergies in children showed a clearer association with phthalates in floor dust than those in adults (Ait Bamai et al., 2014), and that younger children had higher phthalate intake levels than older family members (Ait Bamai et al., 2015). No study has assessed the effect of phthalate exposure, indicated by daily phthalate intake estimated from both urine metabolite and house dust levels, on the risk of allergies in children. The aim of this study was to investigate the association between allergies and phthalate exposure in Japanese children aged 6–12 years old, estimating daily phthalate intakes through both urinary phthalate metabolite level and the phthalate levels in house dust collected from surfaces at floor level and above.

2. Materials and methods

The study design and methods are described in detail elsewhere (Ait Bamai et al., 2013; Ukawa et al., 2013); therefore, only brief descriptions are provided here.

2.1. Study population

The study enrolled 184 elementary school children from 128 families in Sapporo, Japan. The study was conducted in two phases: a baseline questionnaire survey conducted in 2008, and a questionnaire and environmental measurements survey conducted in October and November of 2009 and 2010. The details of the baseline questionnaire survey in 2008 have been reported previously (Ukawa et al., 2013). Briefly, 6393 school children from 12 public elementary schools in Sapporo were asked to participate in the study. Of these, 4408 children responded to the questionnaire (response rate 69%). A total of 951 children (832 families) agreed to a home visit to conduct environmental measurements. After excluding those who had omissions on the baseline questionnaire regarding gender, school grade, and allergies, and those who already graduated from elementary school at the time point of home visit, we contacted the parents of 681 children in 2009 and 2010. As a result, 128 families were available for home visits for environmental measurements (80 in 2009 and 48 in 2010). This selection procedure identified a total of 184 children from 128 families who completed a questionnaire survey in October and November of 2009 and 2010.

2.2. Questionnaire

Self-administered questionnaires were distributed and collected by the investigators during the home visit. The questionnaires consisted of two sections concerning personal and dwelling characteristics. The personal questionnaire gathered information on gender, age, school grade, current body weight and height, history of allergies, parental history of allergies, and household income. Parental-reported allergies were defined by using the International Study of Asthma and Allergies in Childhood (ISAAC) core questionnaire (The ISAAC Steering Committee 1998). Wheeze was defined as “Having wheezing or whistling in the chest in the past 12 months.” Allergic rhino-conjunctivitis was defined as (a) “In the past 12 months, having a problem with sneezing or runny or blocked nose when he/she did not have a cold or the flu,” or (b) “In the past 12 months, having this nose problem with accompanying itchy watery eyes.” Atopic dermatitis was defined as (a) “Having an itchy rash that comes and goes for at least 6 months,” or (b) “Having the aforementioned itchy rash at any time during the last 12 months,” or (c) “Having the aforementioned itchy rash that affects one or several of the following areas: the folds of the elbows, behind the knees, in front of the ankles, under the buttocks, or around the neck, ears, or eyes.”

The dwelling questionnaire included questions about type of building (single-family house/multi-family house); building structure (wooden/other); building age; renovation; floor materials (PVC/compressed wood/wall-to-wall carpet/other); smoker at home; usage of mechanical ventilation system for 24 h (yes/no); dampness-related problems such as condensation (yes/no), moldy odor (yes/no), visible mold (yes/no), high humidity in the bathroom (yes/no), and water leakage problems within past 5 years (yes/no).

2.3. Measurements of phthalates in settled dust

The dust collection methods, gas chromatography/mass spectrometry (GC/MS) analytical methods, quality control and quality assurance, and dwelling characteristics investigations have been described in detail previously (Ait Bamai et al., 2013; Kanazawa et al., 2010).

Briefly, dust samples were categorized as either floor dust or multi-surface dust. Floor dust samples were collected from the living room floor surface and from objects < 35 cm above the floor, such as the carpet and skirting board. Samples of multi-surface dust were collected from the surfaces of objects that were > 35 cm above the floor, including shelves, cupboards, moldings, frames, TV sets, and interior materials such as the corner of the walls and ceiling. The same type of handheld vacuum cleaner (National HC-V15, Matsushita Electric works, Ltd., Osaka, Japan; 145W) equipped with a paper dust bag (Nichi-nichi Pharmaceutical Co., Ltd., Mie, Japan) was used at all dwellings. Background phthalate levels of the vacuum cleaner and the filter were examined and confirmed to be negligible.

The collected dust was weighed after the removal of unwanted substances, such as human and animal hair, food scraps, scrap paper, etc. Samples were stored in stoppered glass test tubes that were sealed with fluoric-tape, wrapped with aluminum foil, and kept at −20 °C until analysis.

GC/MS in SIM mode was used to analyze phthalate levels from collected dust. The target compounds were: dimethyl phthalate (DMP), DEP, di-n-butyl phthalate (DnBP), DiBP, BBzP, DEHP, and DiNP. The analyses were conducted at the Tokyo Metropolitan Institute of Public